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HEALTH PHYSICS DIVISION

PARTICULATE AIR CONTAMINATION AT ORNL

FOR THE DIRECTOR, ORNL

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Authors: J. S. Cheka H. J. McAlduff  
Abstract: Particulate contamination was observed from slug ruptures in the graphite reactor. This report describes monitoring done to characterize particulate released, including during RaLa runs and I-131 cycles. Dust control/removal equipment added in 1948. Contaminated areas grassed and paved.  
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HEALTH PHYSICS DIVISION

PARTICULATE AIR CONTAMINATION AT ORNL

by

J. S. Cheka and H. J. McAlduff

AUG 25 1949

June 30, 1949

O A K R I D G E N A T I O N A L L A B O R A T O R Y

operated by

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Particulate Air Contamination at ORNL

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Preliminary Investigation

a. First Phase

Surveyors at 105 Building had previously reported that localized spots of significantly high activity had been noted in the vicinity of the reactor building and fan house. It seemed logical, then, to use this area as the starting point for the investigation. Scanning of this vicinity with a Victoreen 263 (GM detector) indicated that localized spots of high activity, such as previously noted, could frequently be found. In most cases these could be shown to be due to one, or a few discrete particles. These results suggested that the reactor was a probable source of particulate contamination.

Furthermore, during the removal of ruptured slugs from the reactor matrix, it had been noted that visible clouds of dust were swept from the channels by the air when the slugs were moved. Consequently, an oil-soaked, cloth-covered probe was inserted into the effluent air duct to capture any entrained particulates by impingement. On removal after four days the probe read 200 mr/hr at 1.5" by means of a QT  $\pi$  (thru side wall of chamber). This activity decayed to 20 mr/hr in 24 hours. Washing in  $\text{CCl}_4$  removed large numbers of particulates, both active and inactive, which had impinged on the cloth. Thus, it had been established that the ground surface was con-

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taminated by radioactive particles, and that some, at least, were being discharged by the reactor. However, the extent of surface contamination, the rate of discharge of particles from the reactor, other possible sources of particulate contamination, and the nature of the particles were still unknown. Although it was not believed that a serious condition existed, it seemed desirable to put forth a concentrated effort on the problem in order to secure an early answer to the questions raised<sup>(2)</sup>.

On June 17-19, 1948, teams of Health Physics surveyors, equipped with Victoreen 263's scanned several areas on the plant site, comprising a total of 7500 ft<sup>2</sup> and ranging in distance from 300' to 1675' west and northeast of the reactor stack. From these areas a total of 108 samples of localized activity were collected, yielding 76 discrete radioactive particles. Samples consisting of dispersed activity were discarded. The particles isolated showed similar characteristics, viz., they were dark in color, appeared crystalline, and showed both  $\alpha$  and  $\beta$  activity. These operations supplied a source of particles for qualitative study.

The particles were checked for size,  $\alpha$  activity,  $\beta$  and  $\gamma$  activity, decay rate, and spectrographically for chemical constituents. Sizes of the isolated particles ranged from  $\sim 80 \mu$  to  $400 \mu$  in diameter.

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The alpha activity, uncorrected for self-absorption, ranged from 0 to  $\sim 40$  c/m. Beta-gamma activity ranged from 0.05 to  $\sim 5$   $\mu$ c per sample. However, the lower limit was thought to be determined by the limit of sensitivity of the scanning instrument. Spectrographic analysis showed the presence of U, along with Al, Ca, Mg, Fe, and Si. Presence of the latter elements was considered neither surprising nor significant, since soil usually adhered to the specks. Fission products, though suspected to be present, were not expected to be in sufficient quantity for spectrographic determination. An attempt was also made to determine frequency of fall by the use of sedimentation frames. Results of scanning these with the Victoreen 263's were rather inconclusive.

By the early part of July 1948, it had been shown that: radioactive particles could be found almost anywhere in the plant area, that most of the particles found were too large for inhalation, that they had both  $\beta$  -  $\gamma$  and  $\alpha$  activity, and that uranium was present, indicating the reactor to be at least one source. The latter contention was borne out by the similarity of the particles gathered from the field to those collected in sampling the reactor air duct. Such items as: extent and density of existing surface contamination, rate of new contamination, size distribution, specific activity, chemical and physical nature of the particles, and the resultant potential health hazard

were still undetermined. In view of the fact that it called for an investigation rather than a survey, the problem was turned over to the Special Problems Section of Health Physics.

Meanwhile, although in the opinion of several recognized authorities in the fields of Radiation and Medicine a definite hazard to health did not exist, the necessity for corrective measures, assuming at least a potential hazard in the absence of absolute proof, was recognized. Accordingly, Laboratory management early in August 1948, instituted a full scale program for the elimination or control of radioactive particulates on the X-10 area. This included the design, procurement, and installation of dust removal equipment, the grassing of exposed areas and paving of thoroughfares to immobilize already settled particles, and the supplementing of Health Physics Division's investigation by members of other divisions.

b. Second Phase

A more intensive study was first made of the nature of the particles. While a few particles had previously been submitted to others for various analyses, it was now decided to have the same groups of particles analyzed successively by different groups for different characteristics. Therefore, a group of specimens was sub-



mitted, first to M. Bredig for identity of crystalline structure by X-ray diffraction, then to L.B. Farabee for Pu analysis, then to G.W. Leddicotte for fission product analysis. The results of these, and a few less complete previous tests appear in Table I.

Unfortunately, size measurements and decay tests were not made on these specimens, so that specific activity and "age" could not be simultaneously estimated. However, it does appear that all the particles which showed crystalline structure, whether collected in the field or extracted from the reactor air duct, consisted of  $UO_2$ ; also that all samples analyzed showed a high percentage of Pu, the weighted average being  $88.9 \pm 4.2\%$  of the alpha activity. Fission product distribution showed a greater diversity, but since this is a function of the "age" of uranium, it agreed with the concept that the active particles found were due to oxidation of uranium slugs which had burst their jackets in the reactor. By July 20, 1948, there had been 43 instances of slug ruptures since the first one on September 27, 1944. Some of these were cases of multiple ruptures, an especially severe case having occurred on November 30, 1947.

Specific activity of three particles taken from the reactor exhaust duct was determined. Dimensions were measured by means of a

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Table I

Code	Origin	X-Ray Diffraction Bredig	Pu Analyses - Farabee		Gross $\beta$	F.P. Analyses - G.W. Leddicotte			
			Total $\alpha$ /min	% Pu $\alpha$		Tre	Ce	Sr	Ba
14	500' W. of Stack				8000		4262	1900	790
3A#3	~700' SE		6.5 $\pm$ 0.3	89.2					
4A#5	~500' SW		21.8 $\pm$ 0.6	89.9					
2-1	400' NE	UO <sub>2</sub>	8.9 $\pm$ 0.6	77.5 96.6	8800	2600		1200	1800 1000
1-1	750' W. July 2-12 Frame	UO <sub>2</sub>	1.8 $\pm$ 0.3	94.4 83.3	7350	1800		1200	810 500
PD-A	Reactor Duct	UO <sub>2</sub>	49.0 $\pm$ 1.0	89.0 87.0	69600	41200		5100	2750 2400
PD-B	Reactor Duct	Not Cryst.	5.2 $\pm$ 0.4	100.0 98.1	2300	1500		1000	300
PS	Paint Shop	UO <sub>2</sub> Tr. Graphite	6.8 $\pm$ 0.4	85.3 86.8	7700	4400		435	500

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filament micrometer. The specimens were then dissolved on Pt discs in  $\text{HNO}_3$ . After evaporation, the discs were counted for  $\beta$  -  $\gamma$  and  $\alpha$ .  $\beta$  -  $\gamma$  counts were made at 10% geometry with a mica-window counter. Correction was made for backscatter due to Pt. Alpha counts were made at 51% geometry. Specific activity in  $\mu\text{c}/\mu^3$  was calculated for both  $\beta$  -  $\gamma$  and  $\alpha$  emitting constituents from these measurements. The results appear in Table II below.

Table II

Specific Activity of Activated  $\text{UO}_2$  Particles

Particle	Volume $\mu^3 \times 10^{-6}$	$\beta$ and $\gamma$ c/m (10% geom.)	$\mu\text{c}/\mu^3$	$\alpha$ c/m (51% geom.)	$\mu\text{c}/\mu^3$
12	3.39	6,997	$7.5 \times 10^{-9}$	31	$4.1 \times 10^{-12}$
14	1.47	4,297	$1.06 \times 10^{-8}$	23	$7.3 \times 10^{-12}$
16	3.12	45,612	$5.3 \times 10^{-8}$	44	$6.2 \times 10^{-12}$

Note: A hypothetical particle giving  $\beta$  tolerance\*<sup>(3)</sup> at radius  $40\mu$  contains  $1.3 \times 10^{-7} \mu\text{c}$ .

A hypothetical particle giving  $\alpha$  tolerance\* at radius  $40\mu$  contains  $3.0 \times 10^{-11} \mu\text{c}$ .

\* While true tolerance for a point source is not known, the term as here used indicates a rate of irradiation of 0.1 rep of  $\beta$  per gram of tissue or 0.01 rep of  $\alpha$  per gram of tissue/24 hr.

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The 40  $\mu$  radius was chosen because it is the approximate range of Pu  $\alpha$ , and within this range  $\beta$  and  $\alpha$  irradiations will be additive. The above figures show that the  $\alpha$  exposure due to a 1  $\mu^3$  particle of this nature at 40  $\mu$  is about 0.2 times "tolerance". The  $\beta$  activity on #16 at 40  $\mu$  is about 0.4 times "tolerance". Since the volume, and consequent total activity, of a particle is a function of the cube of the linear dimensions, and the range size for alveolar penetration is considered to extend to 5  $\mu$ , a deeply inhaled particle may have  $\sim$  600 times this total activity of six tenths times "tolerance". Also, since these particles were known to be at least 15 days old, it is likely that a freshly emitted particle would be more active by at least an order of magnitude.

On August 10, 1948, three of the areas previously surveyed (approximately June 18) were again probed with V-263's. This time sampling areas consisted of three 10' x 5' rectangles at each location. Results of both surveys appear in Table III below.

Table III

V-263 Survey

Pt.	Direction From 105 Stack	Description	Particle Density	
			7/18/48	8/10/48
1	825' WSW	Recent fill	5/500 ft <sup>2</sup>	12/150 ft <sup>2</sup>
2	1425' WSW	Roof of 703-A Annex	2/1000 ft <sup>2</sup>	33/150 ft <sup>2</sup>
3	425' NE	Flat grassy	6/400 ft <sup>2</sup>	32/150 ft <sup>2</sup>

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There were seven cases of slug ruptures between these two dates. It may be, however, that the great increase in density of detected particles is not a true measure of fall-out during this period, as the first survey had as its primary purpose the gathering of particles for study, and the check on particle density was incidental. The second survey was made with the express purpose of evaluating the surface contamination. The threshold sensitivity of the V-263 is approximately 400 c/m (at 10% geometry, measured by a mica-window counter), subject to variation due to background caused by less active specks. A few sedimentation frames were placed on the plant area by this time, but since they were also checked with V-263's, not much of significance had been noted. These will be described later.

Decay rates were determined on several particles, both from the field collection and from the exit air duct. Apparent half-lives ranging from a few days to ~ 200 days were noted. In each case, the specimens showing a fast decay rate also showed an increasing half-life as time went on. This is to be expected if one tests particles of irradiated uranium of differing cooling age.

Simultaneously with these qualitative measurements, the problem of the extent and rate of contamination was also attacked. This entailed the use of dust collecting equipment, and an increase in the size of the investigating group.

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Equipment and Techniques

Two phases of the problem of particle distribution were covered by different classes of apparatus. Sedimentation frames were used to determine the rate of fall-out of the heavier particles which contributed to the contamination of the ground surface of the area. Air samplers, mainly filters of various types, were used to ascertain the particle density in air.

The sedimentation frames were 3' x 4' x 5" plywood boxes. These were lined with kraft paper which was held in place by means of a removable 1" quarter-round molding frame. At first a mat of fibre glass was placed over the paper to immobilize settled particles. While the majority of collected dust sifted through this mat, it was later found that a varying fraction was being retained and use of the mats was discontinued. When samples were collected, it was the paper liner which was changed.

The liners were taken into the laboratory, and all dust concentrated onto an area which could be covered by a 14" x 17" X-ray film for radioautography. This technique was later modified by the use of an Airway "Sanitizer" to remove the collected dusts from each paper liner into a removable filter paper bag, which was sealed and radioautographed. The Airway "Sanitizer" is a type of vacuum cleaner differing from others in that it has as a dust receptacle a filter paper

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bag which is removable and replaceable. This modification tended to prevent the loss of part of the specimen by scattering, and also was more effective in collecting that part of the dust which adhered to the paper. It would take seven sheets of film of the above size to cover a paper liner without some method of concentration.

The Sanitizers were also used as direct air sampling equipment. The machines were placed on 3' metal stands, to prevent the exhaust air stream from stirring up settled dust, and run without hose or floor nozzle. The filters were periodically changed, sealed, and radioautographed.

The air is drawn up by a high pressure 2-stage turbo fan, and the flow is subject to variation with filter resistance. The capacity is rated at 65-70 cfm with a clean filter bag and support pad. Velometer measurements showed a flow rate varying from  $\sim 70$  cfm to  $\sim 35$  cfm depending on the degree of the loading of the filter. For purposes of calculations the figure of 56 cfm was used. G.J. Borkowski found the bag to have a surface density of  $12 \text{ mg/cm}^2$ , and estimated the efficiency to be  $> 95\%$  for particles of  $1 \mu$  or larger<sup>(4)</sup>. It was also estimated that a particle  $> 10 \mu$  would be unlikely to be deflected into the nozzle of the machine.

Figure I illustrates the sedimentation frames and the Sanitizers.

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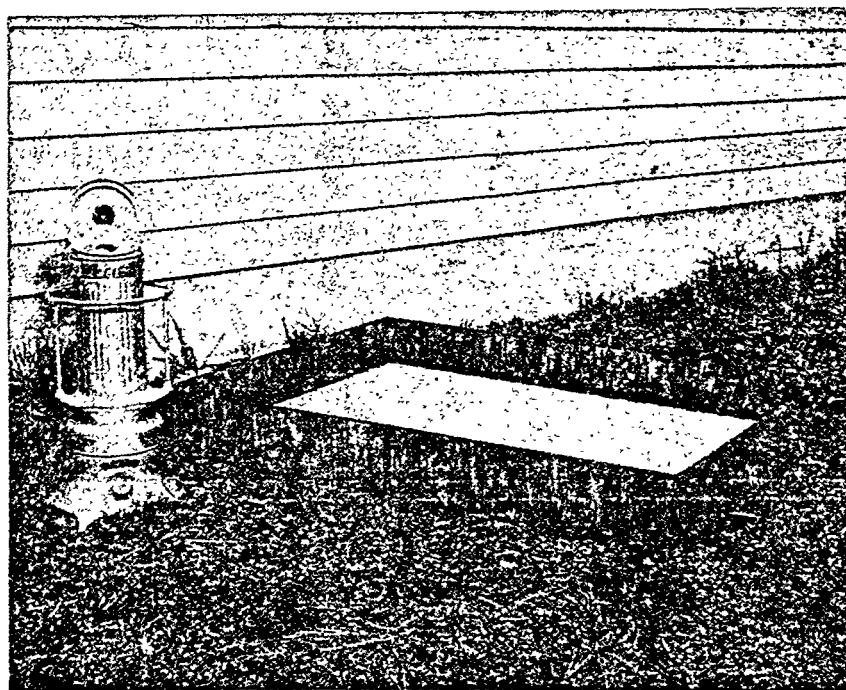


FIGURE I  
SEDIMENTATION FRAME AND AIRWAY "SANITIZOR"



The filtron was another of the air samplers used. This apparatus consists of a "Sutorbilt" constant displacement pump drawing 5 cfm of air through a cylindrical cartridge lined with a Hollingsworth and Vose 9 mil filter paper. Efficiency figures for this paper are not available but are known to be much higher than those for the Standard Airway Filter bag.

The U.S. Public Health Service supplied us, on loan, five large capacity air samplers. These had a rated air flow of 275 cfm through 34 ft<sup>2</sup> of filter paper. These were modified to take 10 ft<sup>2</sup> of filter paper. GWS #6 was used. This paper has an efficiency of 99.97% for the most penetrating particle size. The increased resistance of the system due to the modification cut the air flow to 240 cfm, as determined with a velometer.

These two pieces of apparatus are illustrated in Figure II.

Several constant air monitors are installed about the plant area, three of which are at outdoor locations. These are similar to the filtrons as regards the air-flow system and the filter. In addition, the filter cartridge is shielded by a lead pig, and a GM tube is fixed at its axis. The GM tube is connected to a recording device. The filters from the outdoor locations were periodically radioautographed along with those of our other filtering devices.

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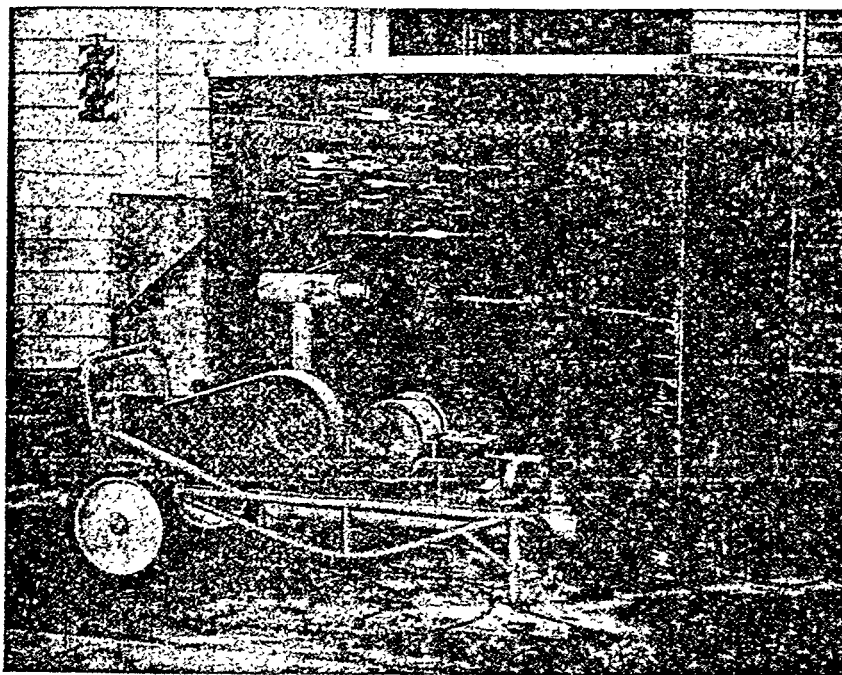


FIGURE II  
FILTRON AND USPHS FILTER

Electrostatic precipitators, drawing 6.5 cfm of air were also tried. These collectors were not satisfactory for continuous monitoring due to voltage breakdown. While intermittent samples could not be directly compared with continuous samples acquired by filters, indications were that the electrostatic method gave us less efficiency and reliability. This method was shortly discontinued.

14" x 17" Eastman "Blue-Brand" X-ray film was used for radioautography. The films were exposed in contact with the specimens, except for the yellow wrappers used by the manufacturer in packing, for 24 hour periods. The wrapper was retained in order to prevent both the contamination of the film and the marring of the emulsion surface. The latter could cause blemishes which may be mistaken for radiation spots. The threshold sensitivity for the method was equivalent to 0.8 c/m at 10% geometry with a mica window counter. When the radioautograph was made through an Airway filter bag the threshold was 3.5 c/m on the same basis.

#### Source of Contamination

Any operation using or processing radioactive material might be a possible source of active particulate air contamination. Some of these have already been mentioned. The reactor, itself, is cooled by ~ 110,000 cfm of air vented through a 200' stack (105 stack), the elevation of the base being 860'. All off-gases from the redox process, and dis-

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solver off-gases from the  $I^{131}$  and RaLa processes are vented through the 200' chemical stack (205 stack) at the same elevation. Airflow through this stack is  $\sim$  20,000 cfm. Cell ventilating air for the RaLa process is discharged through a 50' stack (D stack) north of 706-D building at a flow of 2000 to 6000 cfm. Base elevation of this stack is 820'. Cell ventilating air for the  $I^{131}$  and  $Xe^{135}$  processes is vented through short stacks on the roof of 706-C building.

In addition to these major operations there are the semi-works and several hooded "hot" operations in 706-A. These are vented through short stacks above the roof of 706-A. The incinerator building 626-B, is a possible source of low activity although the state of contamination of burnable refuse is kept at a low level. This stack is  $\sim$  65' high and base elevation is  $\sim$  780'.

Spills at the tank farm cause surface contamination which may become airborne on drying.

The rolling mill, 101-B, may be a source of air contamination of low specific activity in the processing of metallic unactivated U and Th.

Findings (5)

A. Sedimentation Frames


Sedimentation frame coverage had been extended to 17 locations by September 2nd, and to 22 locations by September 15th.

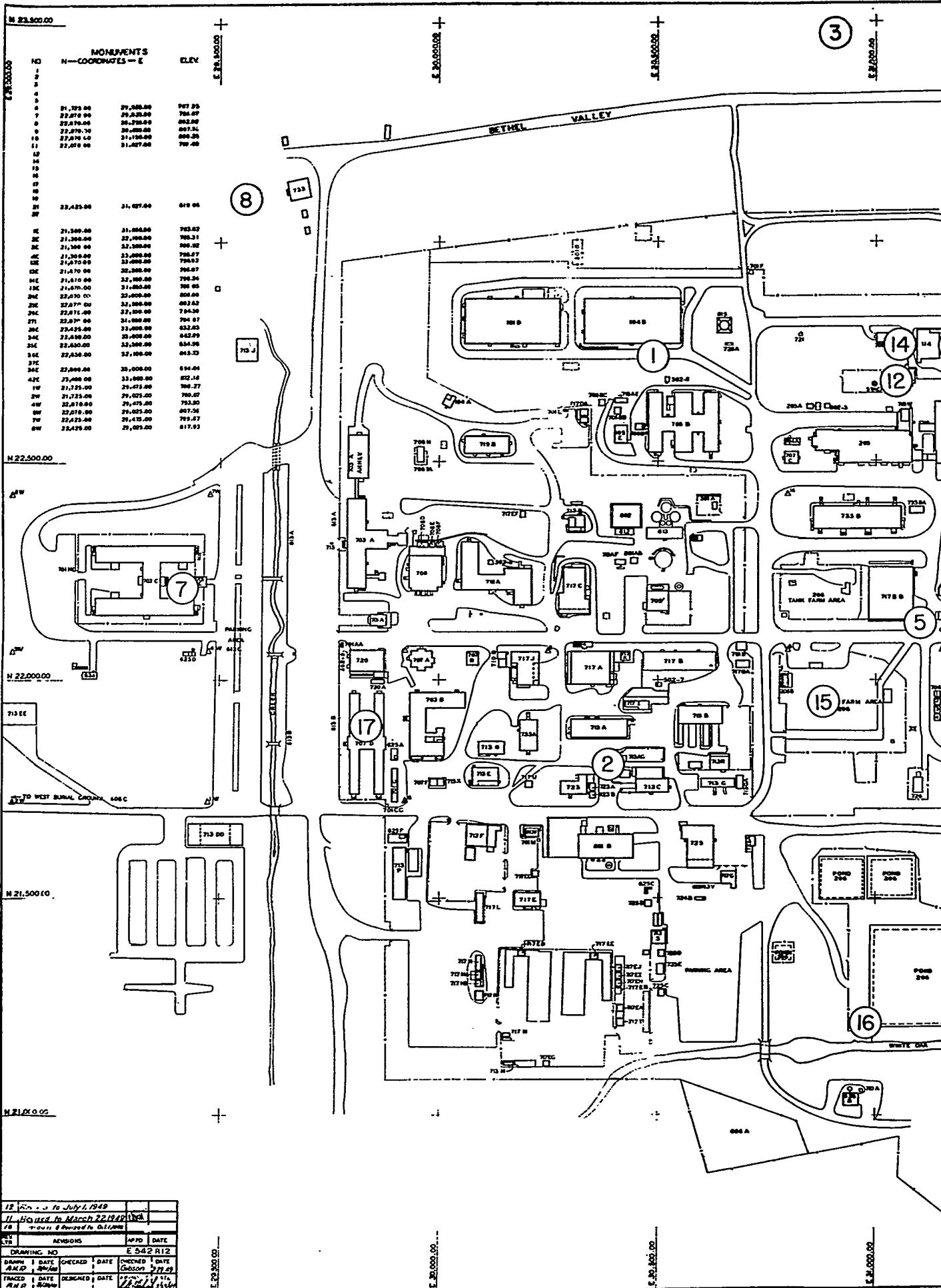
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The final disposition of these collectors is shown in Figure III.

Figures IV, a, b, and c illustrate graphically the frame collections history from August 13, 1948 through February 9, 1949. Group A represents the average value in particles/frame/day of collections from frames within 160' of the reactor stack, group B represents the average of two frames bracketing the 706-D stack at about 300' also about 600' from the reactor stack and group C is the average of all other frames. Daily rainfall is shown in inches by the shaded blocks. Significant events in reactor operation are also shown represented by D for slugs discharged, and R for ruptured slugs, along with the occurrence of the major chemical operations; i.e. the RaLa runs.

It is evident from these figures that the rate of fall-out of active particles in February 1949 is lower by two orders of magnitude than that of September, 1948. It is also immediately evident that there is a sharp temporary drop in collection rate whenever there is rainfall. This may be due to one or more of at least three factors, viz., the rain may remove particles that have fallen on the frames, it may have a scrubbing effect on the air itself, and it may prevent the relocation of previously settled particles.





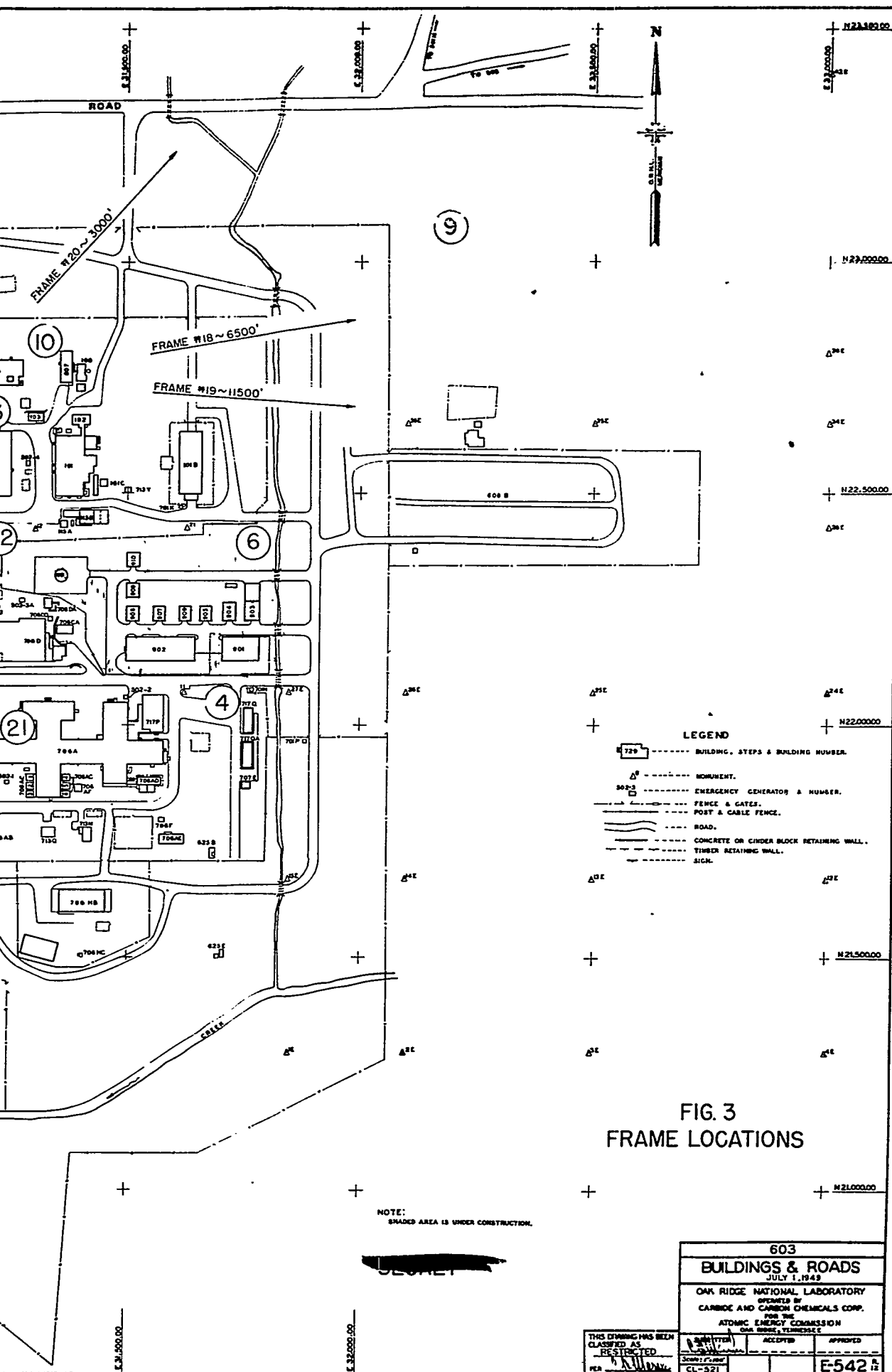
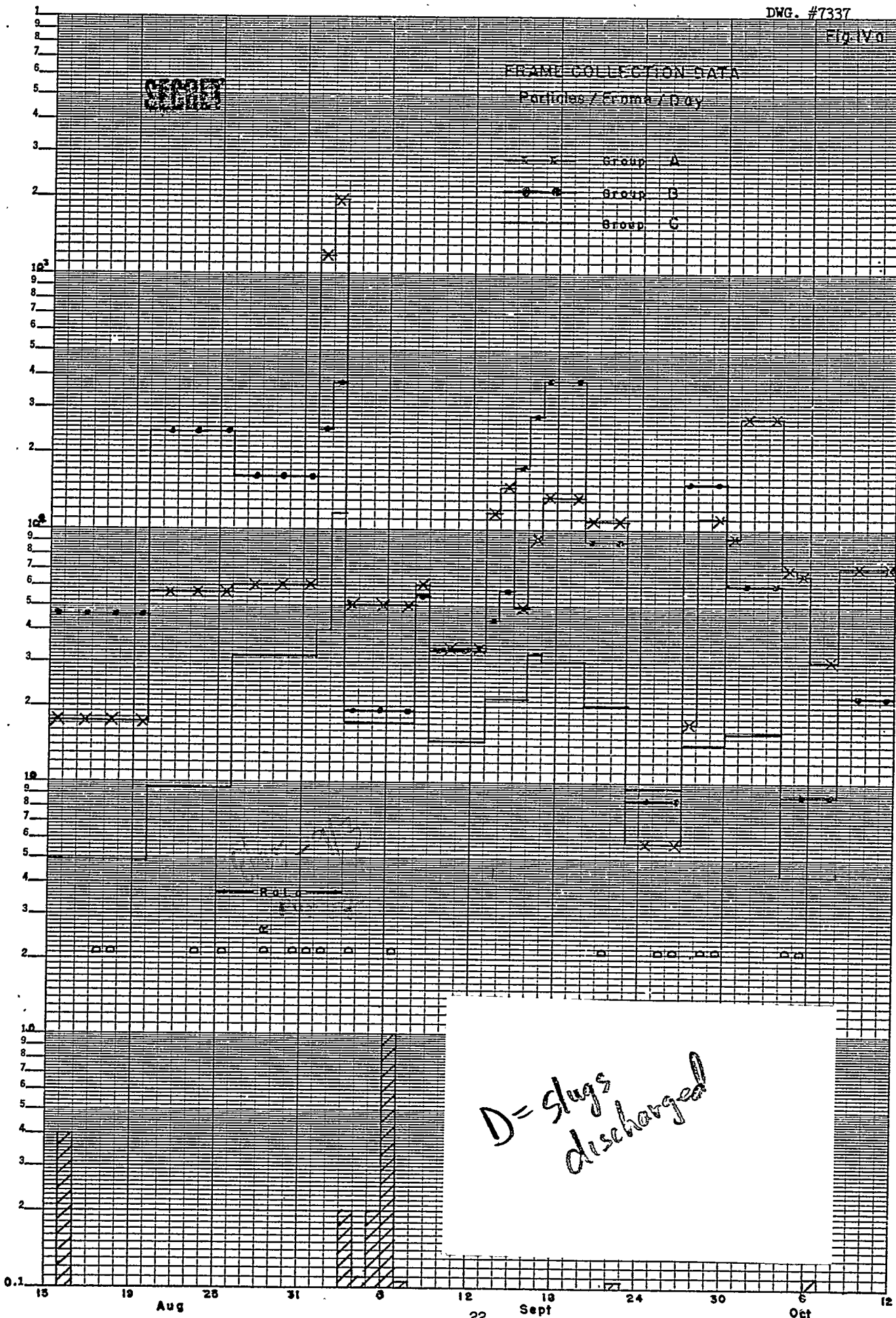


FIG. 3  
FRAME LOCATIONS

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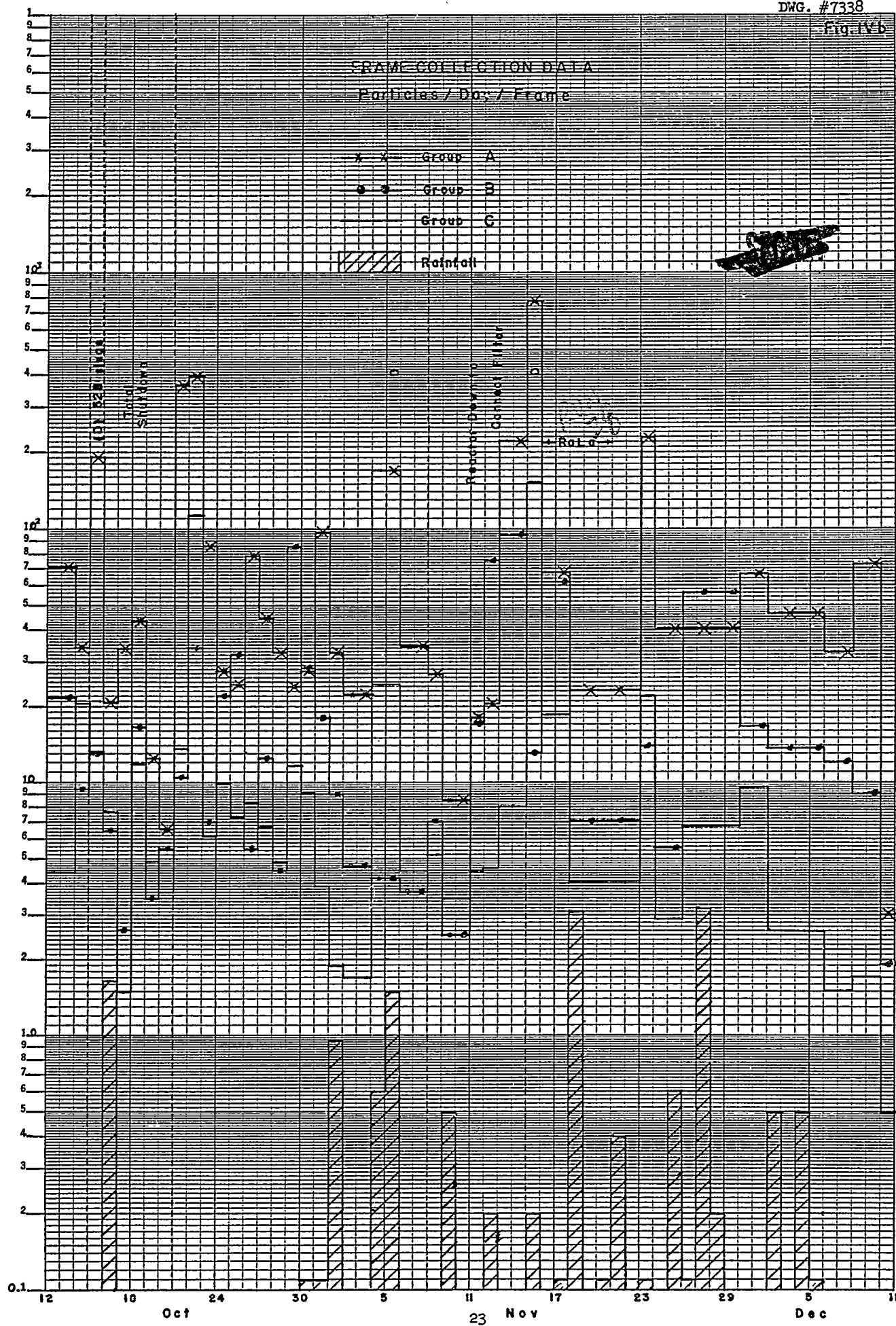
FRAME COLLECTION DATA

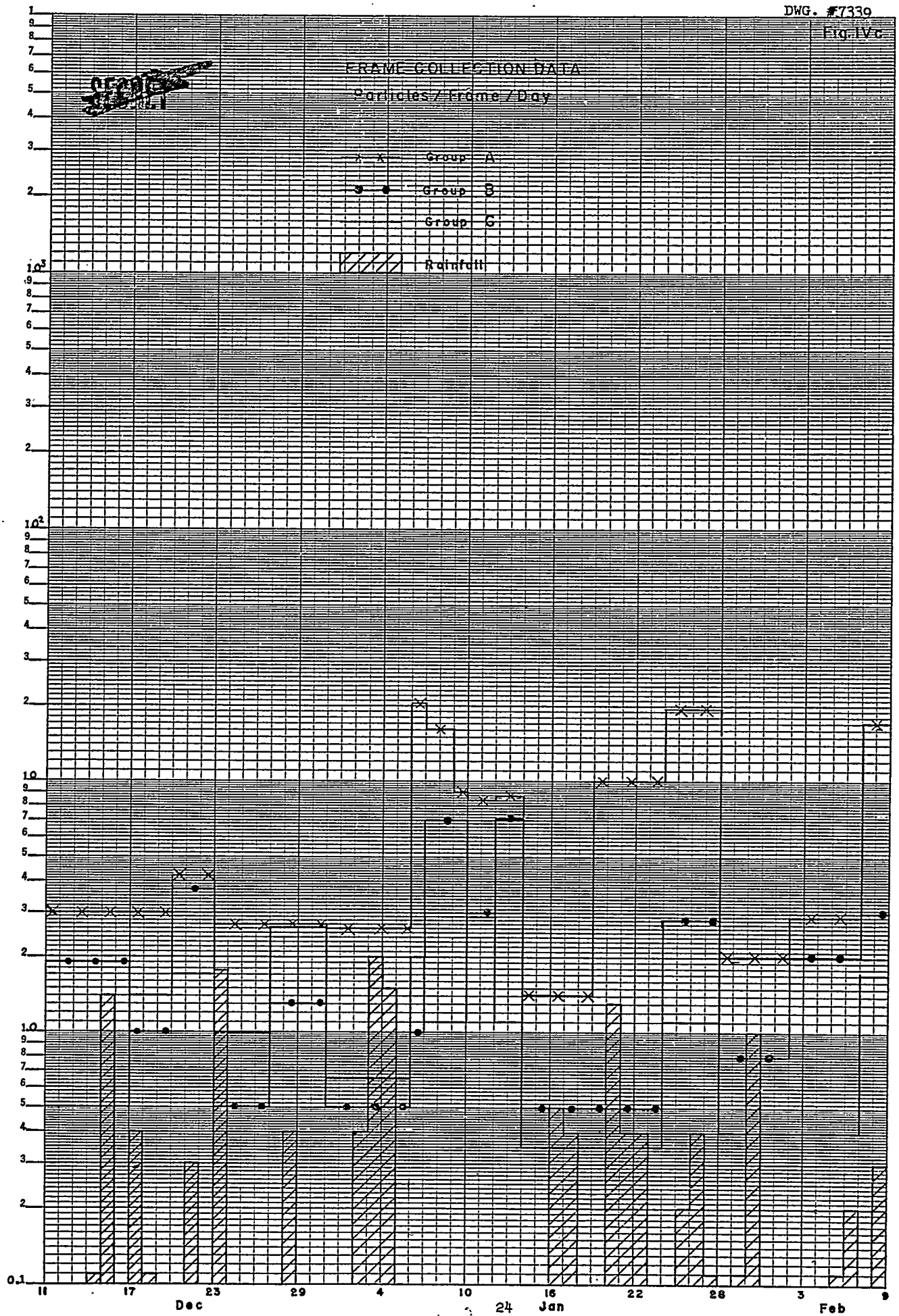
Particles / Frame / Day



*D = slugs discharged*







The largest particle density recorded occurred when the reactor was started up on September 2nd after the removal of the multiple rupture on August 30. It also appears that any disturbance of the reactor channels, such as the discharging of slugs, releases active particles. This implies that the entire reactor matrix is quite contaminated by previously occurring ruptures. The total shutdown of all "hot" operations from October 15th thru 21st caused an overall drop of a factor of four, though the greatest drop occurred near the reactor stack and the 706-D building. The shut-down of reactor operation during November 8-14 to connect the reactor coolant air duct thru the filter house also caused a drop in frame collections. The second highest peak of the survey occurred when this connection had been made, and the coolant air passed thru the filter. While this seems anomalous, it can be explained by the entrainment of pulverized contaminated concrete, crushed during the duct reconstruction, by the coolant air after having passed the filter. The appearance of this group of particles suggested discolored concrete, decay studies indicated half-lives of  $\sim 120$  days rather than about 30 to 60 days as determined from previous frame collections. As might be expected, within a few weeks collection rates dropped to the lowest levels of the period studied. It must be noted, in this connection, that rainfall was both frequent and heavy in this interval, so that relocation was minimized, and total collections dropped, by December to about 1/10 the values noted during total shut-down in October when only secondary sources were operative.

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Group B collections do not follow reactor operation as closely as either group A or C. It was found that collections in this vicinity were somewhat affected by chemical operations, e.g., there was usually an increase in particle count during or after a RaLa run in 706-D or I<sup>131</sup> run in 706-C.

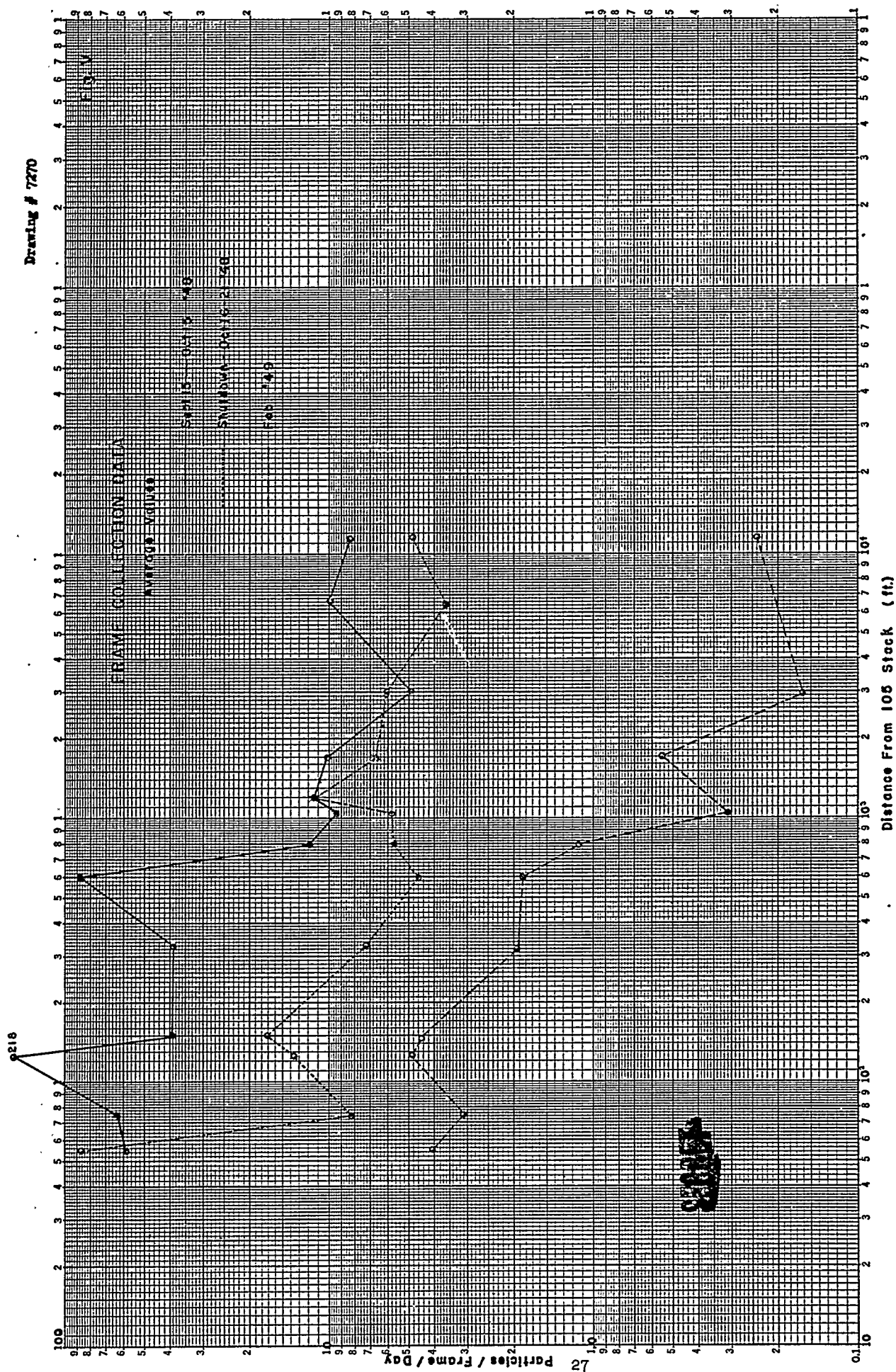
It should also be noted at this time that due to early evidence that relocation was a large contributor to air borne particulate activity, a program of paving all traveled throughfares in the plant areas, and a grassing of all bare spaces was undertaken. This work was begun early in October, and largely completed by the early part of December.

Figure V illustrates the distribution of frame collections with respect to distance from the reactor stack. Three periods are shown: the month between September 15th and October 15th, the shut-down from October 16th thru October 21st, and the month of February 1949. Points represent results from single frames in some cases, but represent the averages of two or more collectors in some cases where the distances were nearly equal.

Several points of interest immediately appear. February collections are down by almost two orders of magnitude from the September-October results. Total shut-down affected near-by values much more than those over 1000' away. The point at 600', representing the frames

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Drawing # 7270



shown by curve B in the previous figure, shows an anomalous high value in the September-October data. This bears out the contention that the chemical operations are also a source of particulates.

Table IV shows these data in condensed form, with time grouping adjusted to include significant periods rather than the calendar dates.

Table IV  
Sedimentation Frame Data  
Particles/Frame/Day

Date	Group A	Group B	Group C	Group D	Remarks
9/1-15	276.4	71.8	37.0	16.8	Removal of multiple rupture
9/16 - 10/15	88.4	90.1	21.6	9.1	
10/16-21	28.9	7.1	6.3	6.3	Total shut down
10/22 - 11/8	108.2	17.1	8.1	10.3	
11/9-13	23.4	22.9	7.3	4.9	Reactor down
11/14-30	116.4	46.9	26.0	19.2	Filter in
December	27.3	11.3	2.5	1.9	
January	10.4	2.3	1.2	0.8	
February	5.4	1.9	1.3	0.4	

Group A comprises the five frames within 170' of 105 stack.

Group B comprises the two frames bracketting 706-C and D buildings.

Group C comprises the six frames beyond group A but less than 1000' from 105 stack, except group B, which is about 600' from it.

Group D comprises the nine frames at more than 1000' distance, maximum 11,500'.

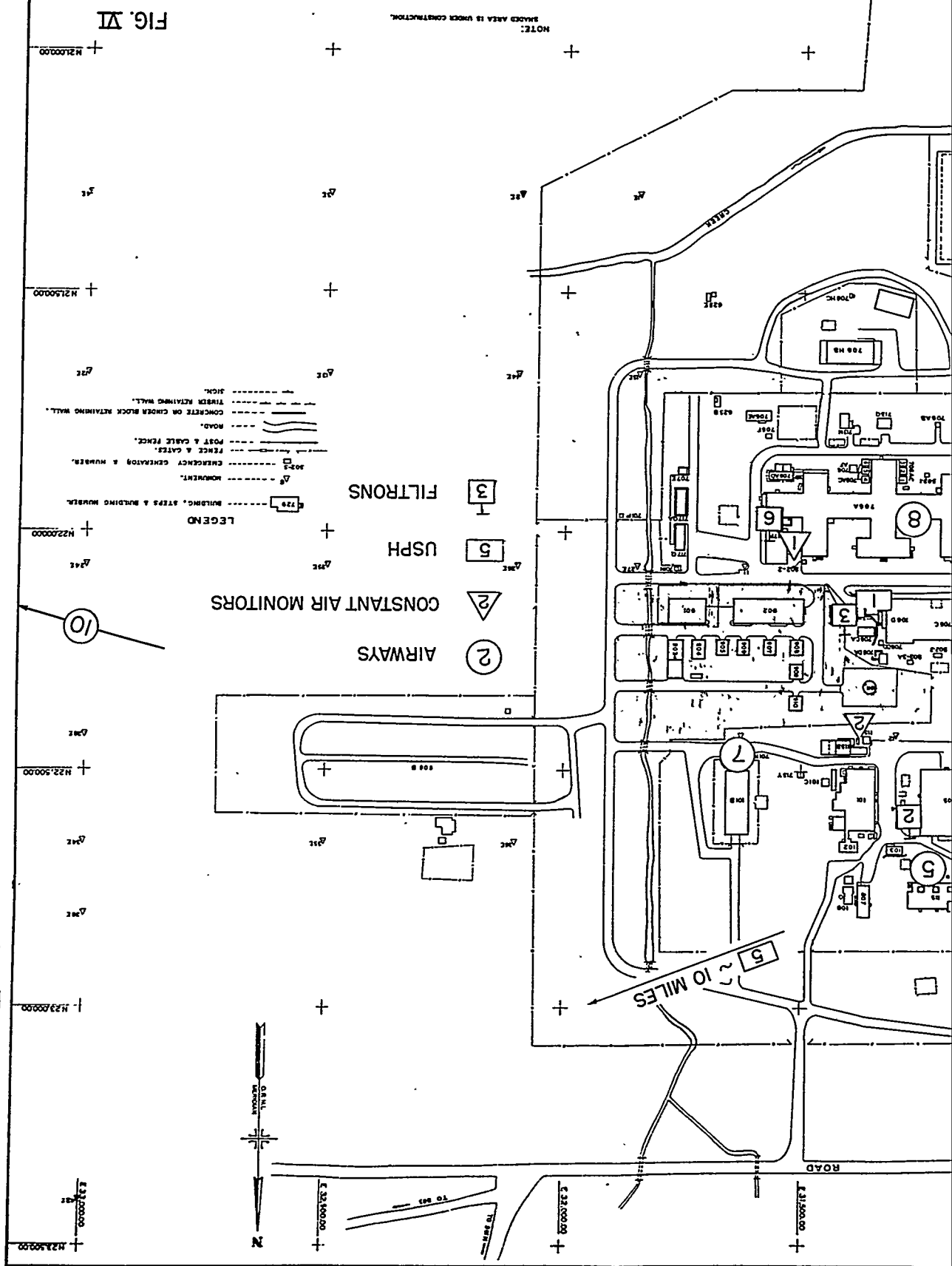
#### B. Filter Type Collectors

There were six filtrons available for use. They were put into use early in August, and by October 5 were in the locations shown in Figure VI. Of the five USPHS filter, four had been modified by September 24, and placed about the X-10 areas as shown in Figure VI. The fifth one was placed at the AEC Administration Building in Oak Ridge on October 14. Five of the Airways were placed on October 10 and by October 15 the other five of the ten that were used were placed, also, as appearing in Figure VI.

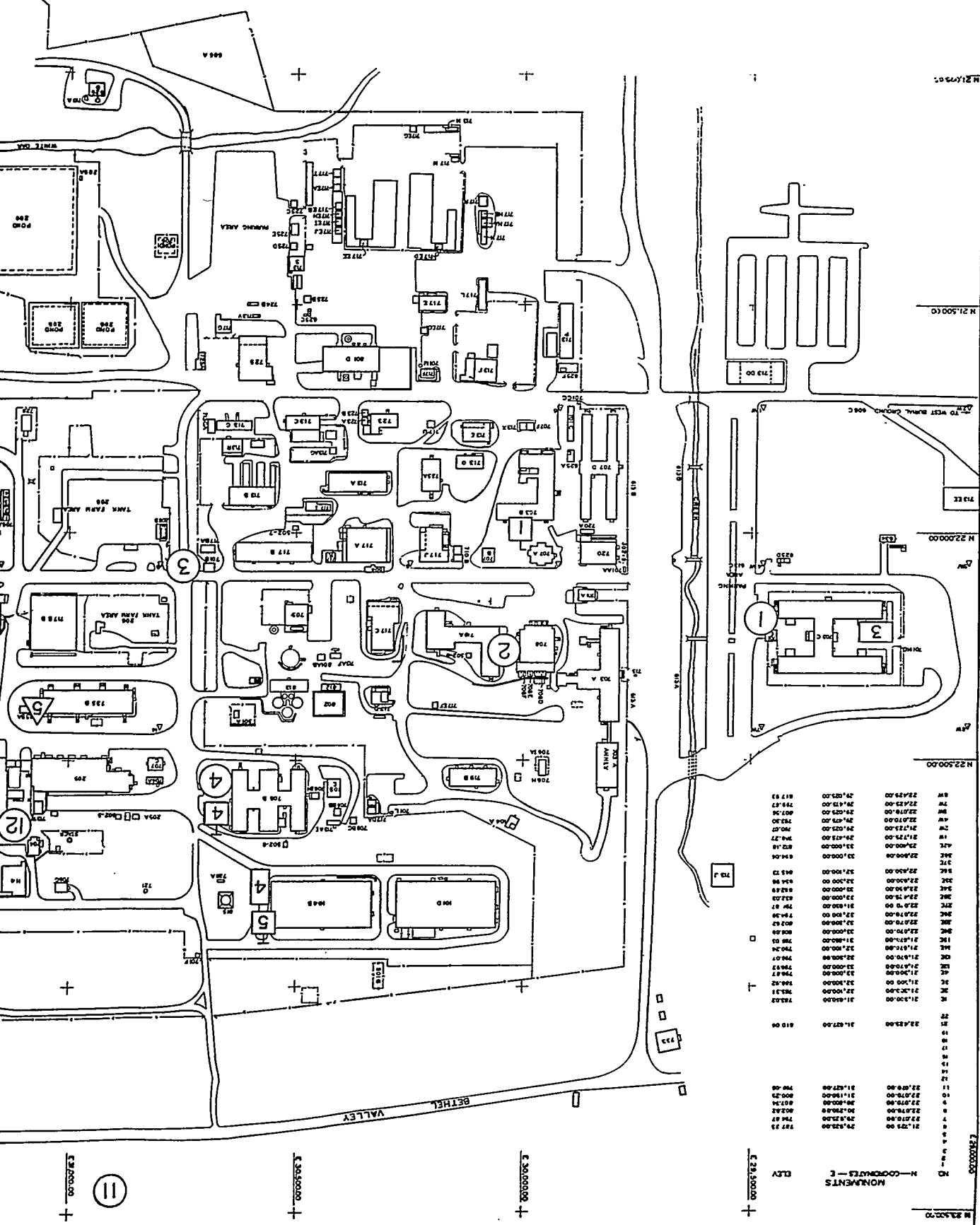
It was evident early in the investigation that filter collection rates did not follow the same trends as frame sedimentation rates. Results could not be compared directly, however, until October 15, since sampling by filtrons previous to that date had been by two or three hour periods while frame collections occurred on a round-the-clock basis.

E-5424		30
THIS DRAWING HAS BEEN REVISIONED DATE 11/19/54 BY 1111/11/54		CL-591
APPROVED AUTHORIZED FOR THE ATOMIC ENERGY COMMISSION CARBON AND CARBON CHEMICALS COM. OAK RIDGE NATIONAL LABORATORY JULY 1, 1949 <b>BUILDINGS &amp; ROADS</b> 603		

FIG. VI



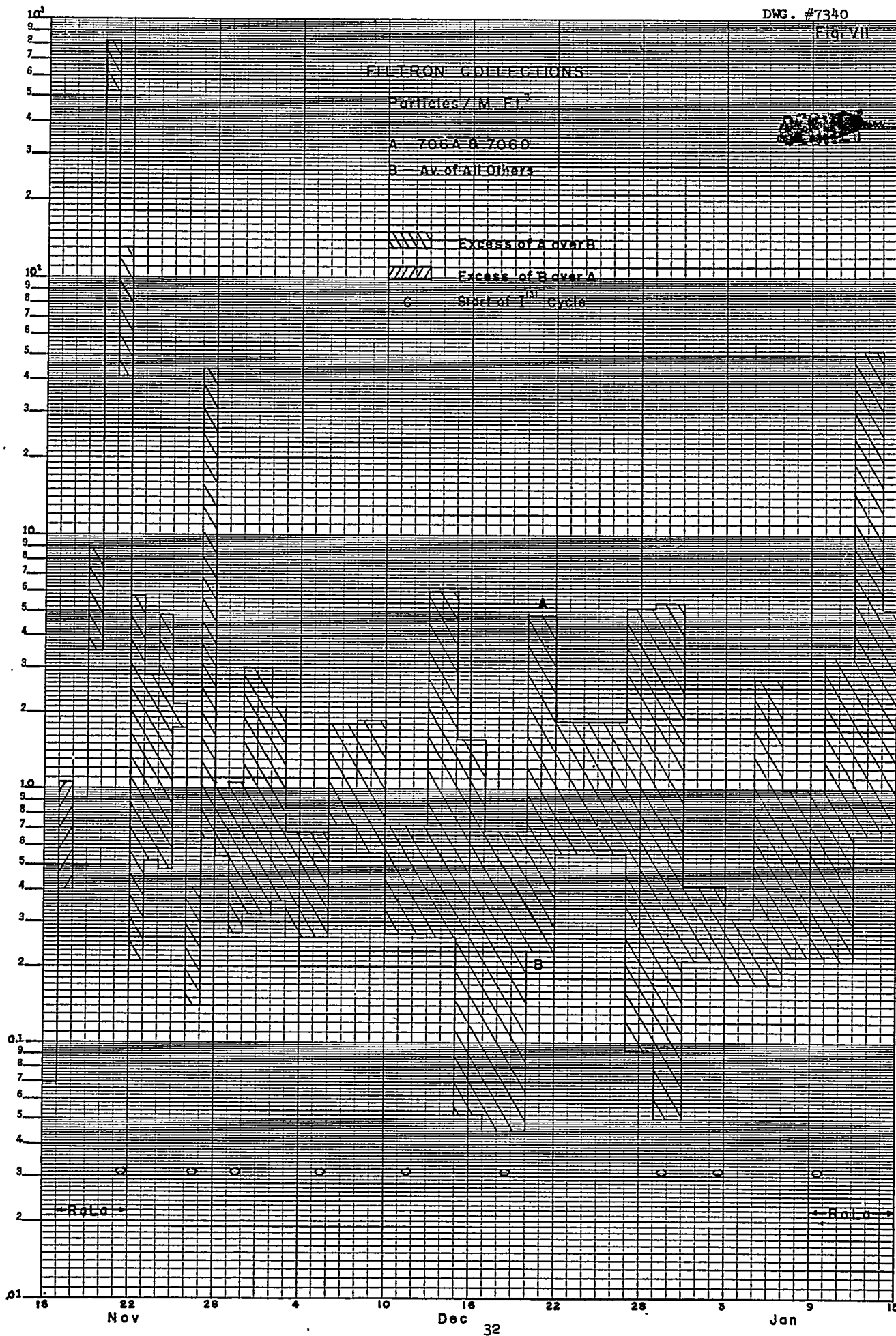


[illegible]

Comparison of data from the six filtrons with frame collection data shows that peaks are only occasionally coincident. It is found that collection peaks occur during the latter parts of the extraction cycles of both RaLa and  $I^{131}$ . This is especially noticeable when one observes the two located just outside of 706-D Building and near the semi-works in 706-A. The sequence is not as closely followed by the more distant stations, but, as the collection rate of filter type devices not within 300' of 706-C and D buildings is usually considerably less than 1 particle/1000 ft<sup>3</sup>, fluctuations are not as significant.

Figure VII illustrates the results of filtron collections during a period of frequent  $I^{131}$  runs, and inclusive of two RaLa runs. Curve A, representing the two filtrons outside of 706-D and A, shows a marked rise within two or three days after each  $I^{131}$  cycle begins (represented by "C"). The extraction cycle usually lasts three days.

It is also interesting to note the effects of the two RaLa runs. The November run was unfiltered. The final evaporation and removal of the product took place overnight, during a temperature inversion. It is quite evident that the cell ventilating air, which is discharged through the 50 foot D stack, was boxed in by the inversion and spread through the whole plant area. A temporary filter had been installed in



the duct venting through D-stack prior to the January run. The rise in general area contamination was almost negligible. However, there was a 50-fold rise in particle count in the immediate vicinity. It appeared from this evidence and other studies which will be discussed later, that airborne contamination was escaping through the building rather than the stack. ✓

The USPHS filters would seem to give most valid figures because of the large volume of air sampled. In one instance (706-D E platform) one of these filters was run near a filtron. In general collection, figures were lower than for the filtrons by a factor of  $\sim 3$ . This may have been due to the fact that the collecting orifice of the filtron is only 2' above floor level, whereas, that of the large filter is  $\sim 5.5'$ . This difference may cause the filtron to draw in more of surface dust which is raised by foot and other traffic. Apart from these, results are not comparable because of differing locations.

The filter at the AEC Administration Building gave valuable information. Collection rates of radioactive particles usually ranged from less than 1 to  $\sim 10$  particles per million  $\text{ft}^3$  of air. The one exception was the 11 day period comprising the November RaLa run, during which time the collection rate was 82 particles/million  $\text{ft}^3$ . The overall average for the three and a half month period from the middle of

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October to the end of January was 9.85 particles/million ft<sup>3</sup>. The average, exclusive of the RaLa run mentioned above, was 1.96 particles/million ft<sup>3</sup>.

It should be noted that none of the activity detected in town had much magnitude. Most of the particles noted were just over the threshold of sensitivity, which, as previously mentioned, was the equivalent of 0.8 c/m at 10% geometry on a mica window counter.

The Airway collections show somewhat different characteristics. First, the radioautographic technique is not as sensitive as when used with other filters because of the interposition of the 12 mg/cm<sup>2</sup> of the filter paper as an absorber. Second, the estimated efficiency of ~ 95% for particles of 1  $\mu$  implies that most of the sub-micron particles may pass through the paper.

The most interesting, and perhaps, significant feature of the comparison between these filters and the filtrons is the change in relative number of particles detected on the two media. The average of all the outside Airway collections compared to the average of the six filtrons gave a ratio of ~ 1:8 in the latter half of October, and nearer 1:60 for December and January. A direct comparison was made at one station (706-B E door). This location showed a ratio of ~ 1:3 during October,

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and again ~ 1:60 during December and January. The greatest difference in general conditions between these two periods was the filtering of the reactor cooling air after the middle of November. It appears from these observations that particulates produced by the chemical operations are of extremely small size though high specific activity, and many possibly pass through the Airway bag. This is further implied by the fact that until the middle of November the Airway collection peaks followed the frame collections much more closely than did the other filter types. Table V gives a summary of collection data from these three types of filters.

Table V  
Particle Data from Filter-type Samplers  
(Particles/M ft<sup>3</sup>)

Dates	Filtrons (6)	USPHS (4)	Airways (10)
10/15-10/31	0.62	0.19	0.08
November (except RaLa)	1.84	0.74	0.15
November (RaLa)	115.7	38.8	7.21
December	1.07	0.39	0.02
January (except RaLa)	1.55	1.08	0.03
January (RaLa)	4.71	11.9	0.05

Note: The RaLa runs in November and January raised particulate air activity so greatly that it was considered more representative of the true picture not to average those values with the balance of the respective months' data.

From this table it is obvious that the improvement evidenced by frame data does not hold in the case of filters. The best aspect of the situation is the fact, as shown by the graph in Figure VII, that the contamination is localized, and that the contamination level of air in the area exclusive of the immediate vicinity of chemical operations is low and falling.

c. Supplementary Studies

Besides the sampling and collection procedures just reported, several studies were made by this and other divisions to attempt to understand more fully the sources and nature of the particulates in question. Some of these were continuations of work mentioned under Preliminary Investigation. Others were suggested during the progress of the project.

The start-up of the reactor subsequent to the August 30 multiple slug rupture coincided with the end of a RaLa run. It was noted that frames near 706-D had a higher active particle incidence than expected from consideration of distance from the reactor stack alone. It was also

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noted that the distribution of activity/particle was much more uniform than on other frames. Figures VIII and IX representing collections from #5 frame near 706-D, and #10 frame 150' from 105 stack illustrate this phenomenon. In addition, the fibre-glass mats, which were still used at this time, retained a larger fraction of active particulates collected near 706-D than near the 105 stack.

To verify the apparent difference in the source and nature of the particles collected at the two locations, a series of tests was made. Specimens from the two locations were first leached with water, and then with  $\text{HNO}_3$ , separating the water soluble and acid soluble fractions. For comparison and identification of sources, a specimen of dust collected by a cloth filter sampling a portion of the reactor cooling air and one collected in the 706-D cell-ventilating air duct by impingement on a filter-paper covered probe received the same treatment. These fractions were allowed to decay, and apparent half-lives observed. The activity in the insoluble residue was negligible in each case.

Table VI is a summary of these tests.



NOT CLASSIFIED  
PHOTO #5081



FIGURE VIII  
#5 FRAME COLLECTION - NEAR 706-D

NOT CLASSIFIED  
PHOTO #3568

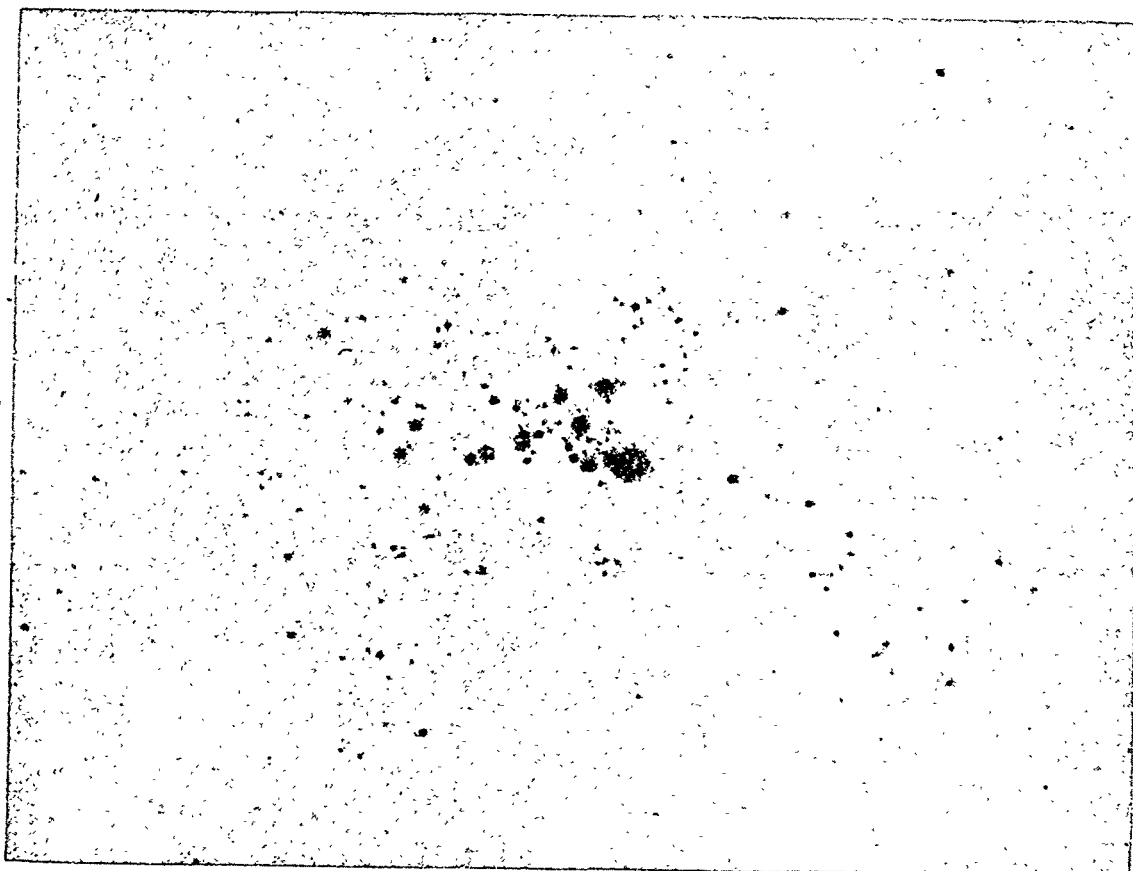


FIGURE IX

#10 FRAME COLLECTION - 150' FROM 105 STACK

Table VI

## Water-and Acid-Soluble Fractions of "Particulates"

Code	Source	c/m (Beta and Gamma)		% of Total	Apparent Half-life (days)
		Water-Soluble Fraction	Acid-Soluble Fraction		
5-W	Near 706-D	3,602		69	37
5-A	Near 706-D		1,606	31	32
5-2-W	Near 706-D	2,396		85	48
5-2-A	Near 706-D		423	15	30
10-W	Near Reactor	855		20	42
10-A	Near Reactor		3,413	80	62
13-W	Near Reactor	1,955		6.7	33
13-A	Near Reactor		27,040	93.3	33
14-W	Near Reactor	406		3.0	45
14-A	Near Reactor		13,084	97	48
J-W	Reactor Duct	9,266		2.4	70
J-A	Reactor Duct		370,000	97.6	70
D-W	706-D Duct	365,000		68	16
D-A	706-D Duct		170,600	32	13

The above data do not show any significant difference in the decay rates of any pair of water-and acid-soluble fractions. What does appear is that the water-soluble fraction of the reactor-produced particles is very small compared to those produced in chemical operations, and the apparent half-life of the reactor-produced particles is longer. Since the

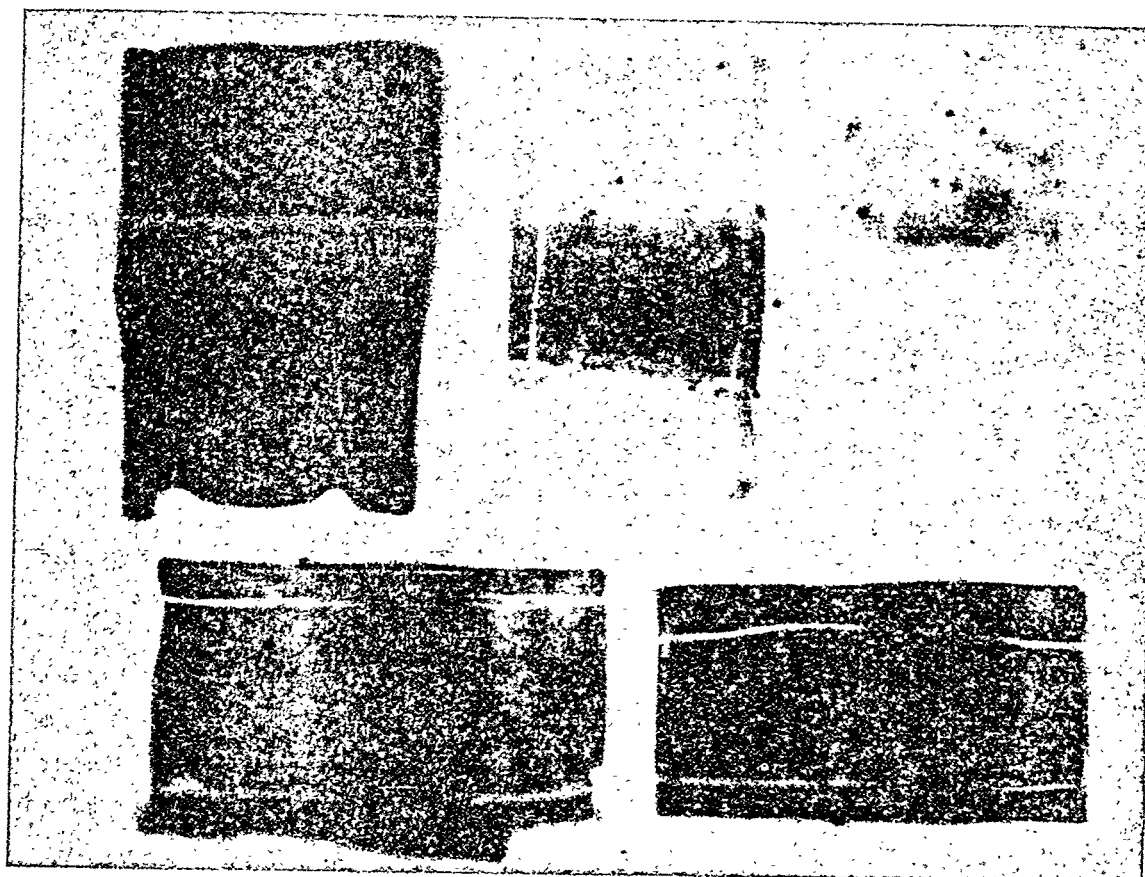


FIGURE X  
FILTERS FROM 706-D DUCT DURING AUGUST RaLa RUN

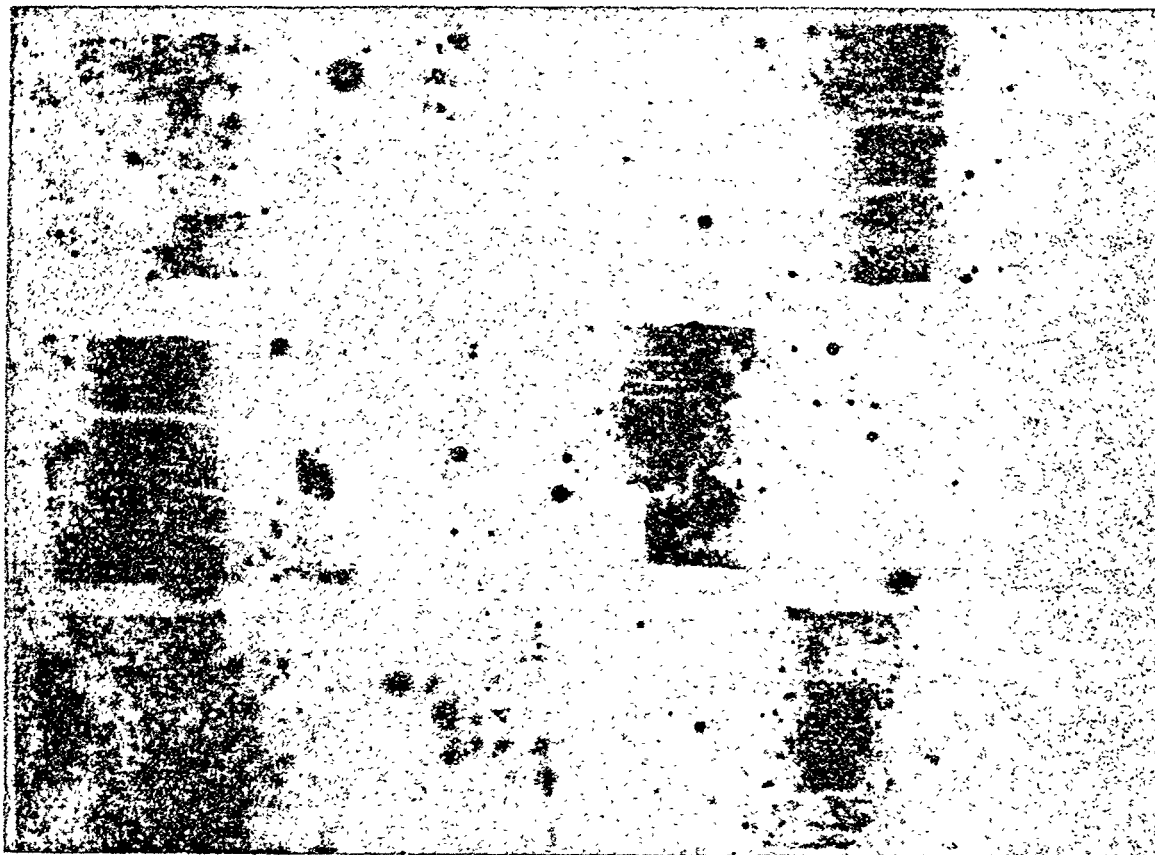


FIGURE XI  
FILTERS FROM 706-D DUCT TWO WEEKS AFTER AUGUST RaLa RUN

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frame specimens from neither location agree closely with the corresponding source in both these characteristics, it may be assumed that field collections are accumulations from both sources in varying proportions.

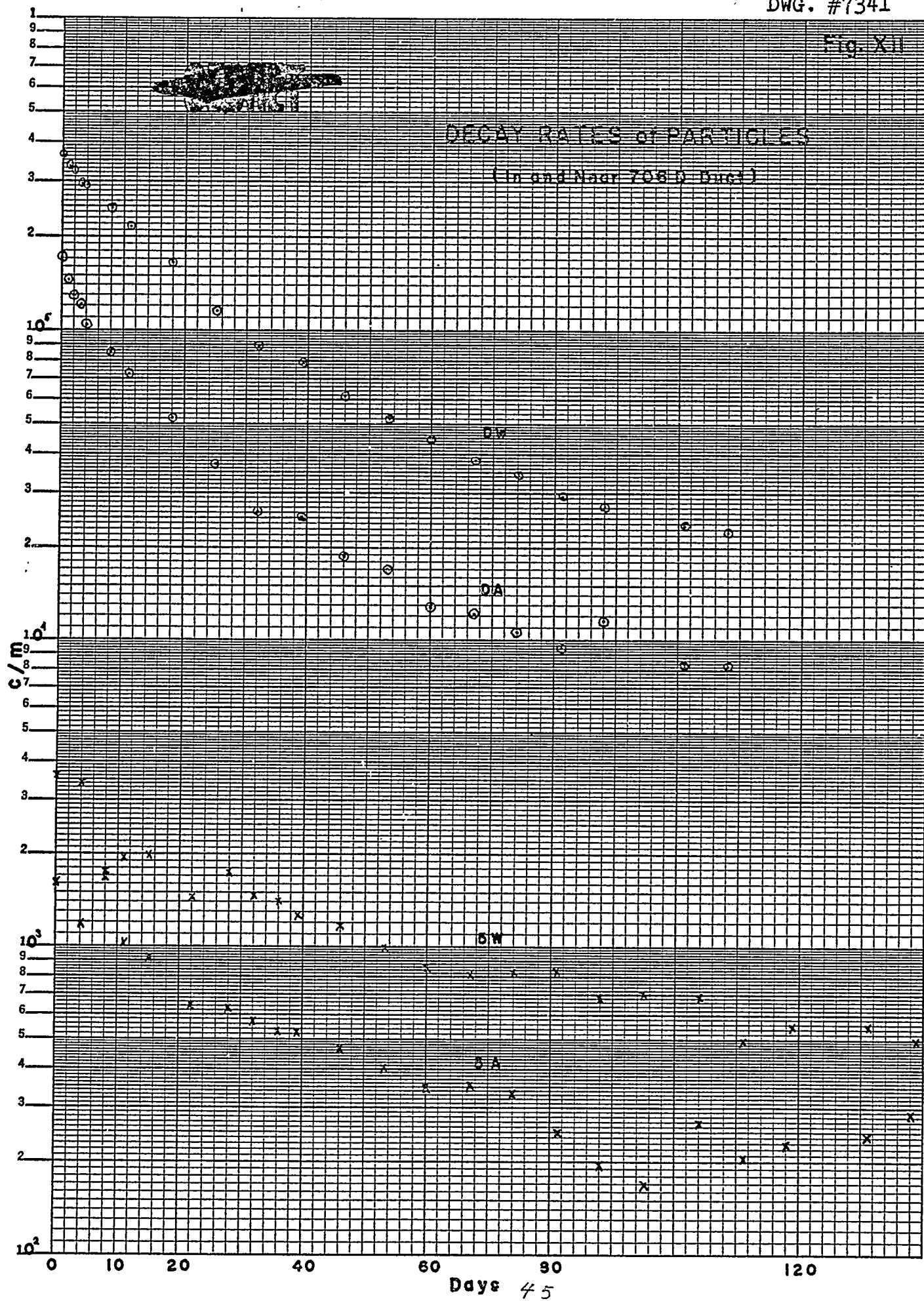
The lack of closer agreement between characteristics of specimens from near 706-D and from 706-D duct is probably due to the fact that fallout from the D-stack effluent is selective. The stack effluent also comprises radioactive material in non-particulate form. That the ratio of particulate to non-particulate material is a variable is shown by Figures X and XI. Figure X shows a radioautograph of a series of filter papers from the probe in the D-duct during the August RaLa run. These were changed daily. The activity shown in this autograph is what is left after one week's decay - complete blackening having occurred on exposure of the freshly removed filters, one of which read 250 mr/hr at 2 inches with a QT  $\pi$  when taken off the probe. Figure XI is a radioautograph of filters off the same probe with daily changes  $\sim$  2 weeks after the completion of the run. The diffuse darkening which appears to a greater or less degree on these radioautographs was also an occasional characteristic of filtron, constant air monitor, and USPHS filters in and near 706-C and D when radioautographed. It was not determined whether it is the same contaminant which appears in different physical form or whether a different contaminant is responsible for the two effects.

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Decay curves for four of the specimens listed in Table VI appear in Figures XII and XIII. From these it appears that in addition to variance in apparent half-lives of the several specimens (shown in Table VI) shortly after collection, the nature of the curves also varies. While all of the curves show long-lived fractions, the sample from D-stack does not exhibit a low decay rate until it has reached less than 10% of its value, while the sample from the reactor duct (J) shows a low rate after  $\sim 50\%$  of its value. The field collections are intermediate, as may be expected.

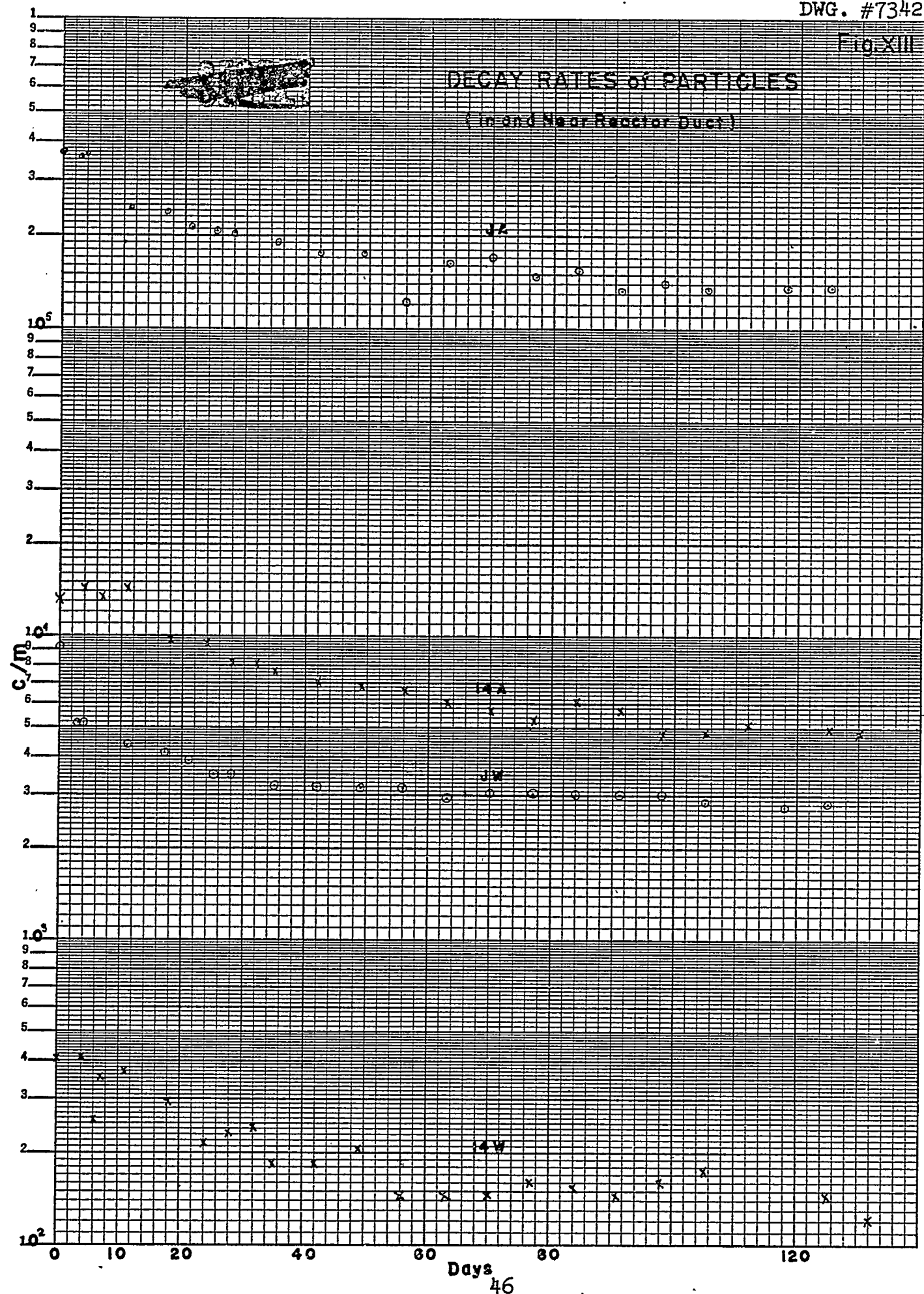
Other decay studies of specimens of air contaminants and sedimentation were made at various times. It was found consistently that samples taken in or near the 706-C and D buildings decayed rather rapidly and had a very small fraction of long-lived material.

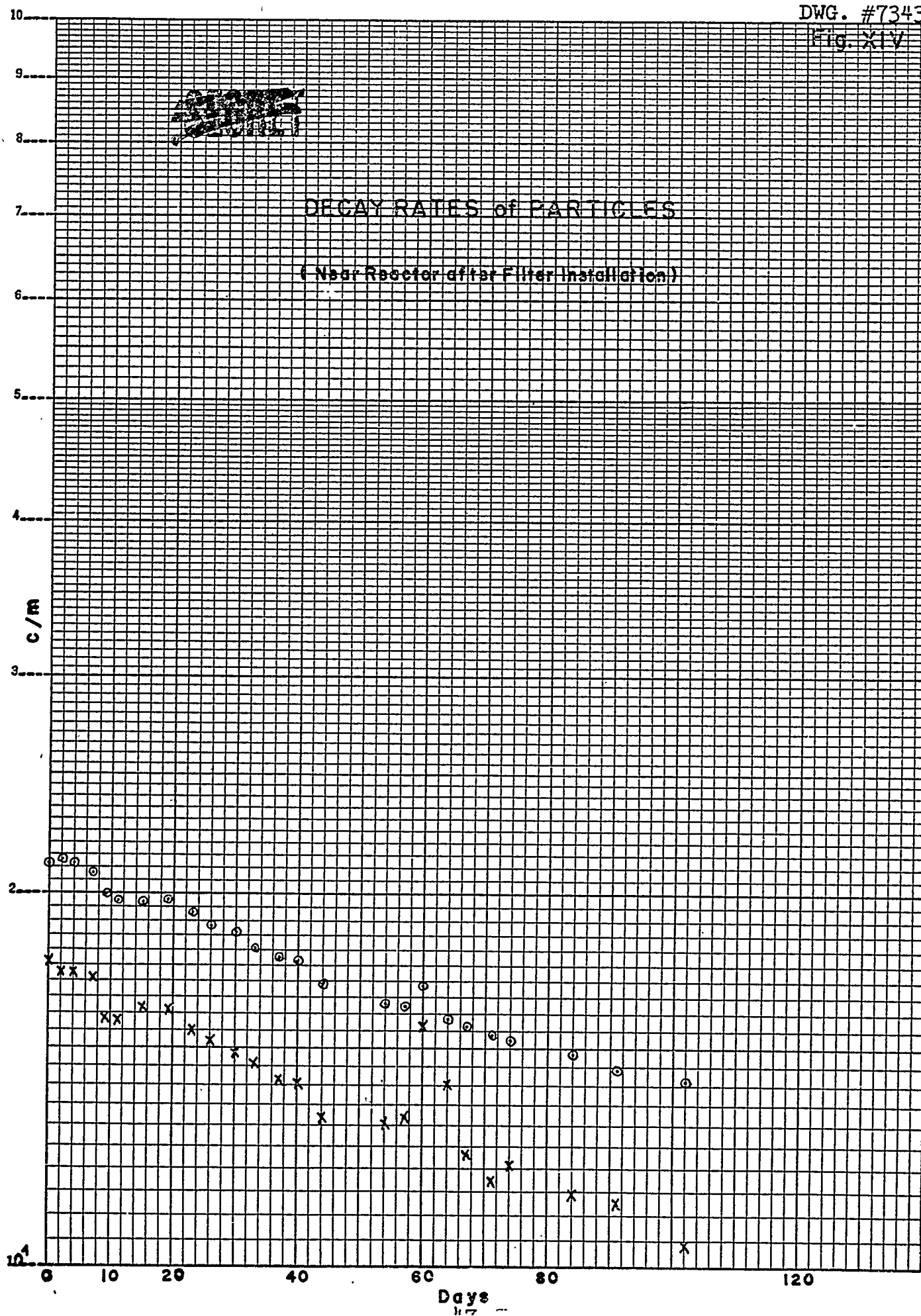
A decay study was also made of sedimentation samples in the vicinity of the 105 stack after the filter had been installed in the reactor coolant air duct. As previously mentioned, there was a large fall-out of active particulates at this time, but their appearance suggested concrete rather than  $\text{UO}_2$ . Figure XIV illustrates the decay pattern of two of these samples. There is apparent in these curves no short-lived fraction. This would seem to establish that all of the activity was due to old contamination of the duct walls dislodged by the construction activity.





# DECAY RATES of PARTICLES (In and Near Reactor Duct)





The "age" cannot be estimated, except that the material is not fresh. A rule of thumb for the estimate of "age" of material, stating that the apparent half-life of a normal aggregate of fission products is a measure of the age, has sometimes been used. This rule cannot be applied to  $\text{UO}_2$  particles issuing from the reactor for two reasons. First, uranium (oxide), irradiated in the reactor in the exposed and finely divided state existing after a slug is ruptured, does not retain a normal aggregate of FP's since many of the fission chains pass through gaseous species which fail to remain totally occluded, and consequently the ends of the chains are lost. Second, even when the aggregates are normal (no loss) the decay scheme of the whole is a function of the time of irradiation as well as cooling time.

The loss of the gaseous fission products was established by R. Livingston of the Chemistry Division, who made a detailed study of the contamination of the reactor coolant air<sup>(6)</sup>. He found that a large fraction of the total activity of the air stream was due to various very short-lived isotopes of Xe and Kr which decay in flight. Decay-products of these gases were collected on a charged wire in his apparatus, and later identified.

P.R. Bell of the Physics Division, also developed a sampling apparatus for the reactor coolant air<sup>(6)</sup>. This consisted of an adhesive

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tape, capturing particulates by impingement and monitored continuously as it left the air duct. This was designed to detect new ruptures by increased particulate activity. Both of these investigations are reported in detail in ORNL-197.

The reactor duct was also sampled with a cascade impactor to evaluate size distribution of activated  $UO_2$  particles. The sampling was done by R.H. Wilson of the University of Rochester on November 4-6. The apparatus used was the University of Rochester modification fitted with a pump drawing 0.5 cfm of air. The separation is produced by passing the air through a series of four jets of decreasing size, causing increasing air speeds, each of which blows against a resin-covered glass slide, thus collecting progressively smaller particles by impingement. A fifth stage was supplied by a filter backing the series.

The instrument had been calibrated for  $UO_2$  (sp. gr. 10.9). It was estimated that the maximum size of particles of  $UO_2$  taken in by the inlet orifice was  $\sim 10 \mu$ . The mean value of the size fractions are:

1st stage	6. $\mu$
2nd stage	2. $\mu$
3rd stage	1. $\mu$
4th stage	0.7 $\mu$
Filter	0.4 $\mu$

It must be noted that if any of the active particulates are other than  $UO_2$  the above distribution is invalid.

The slides were analyzed for uranium content at Rochester by the fluorophotometric method. However, before they were taken to Rochester, several  $\beta$  -  $\gamma$  counts were taken on all samples, and the slides from two of the runs were radio-autographed. Table VII shows the results of the  $\beta$  -  $\gamma$  counts.

Table IX

Beta and Gamma Count on Cascade Impactor Samples

(Figures indicate c/m at 10% Geometry)

Run Number	Mean Diameter (Microns)				
	$>4$	$\sim 2$	1	0.7	0.4
#1a (15 hours after collection)	1372	272	293	176	843
#1b (73 hour decay)	1096	124	81	46	
#2a (1 hour after collection)	987	625	485	309	1763
#2b (73 hour decay)	531	154	88	48	
#3a	4482	337	284	139	233
#3b (7 hour decay)	4533	357	299	123	236
#4a	6779	531	198	76	96
#4b (7 hour decay)	6635	579	201	61	78
#5a	469	388	412	316	4223
#5b (7 hour decay)	175	94	107	41	263

Run #1 was a 7 hour run under normal operating conditions.

Run #2 was a 14 hour run under normal operating conditions.

Run #3 was a 6 hour 19<sup>0</sup> run, reactor down, slugs being pushed.

Run #4 was a 51<sup>0</sup> run, 12<sup>0</sup> of which was start-up of fans.

Run #5 was a 56<sup>0</sup> run, power on.

Runs 1, 2, and 5 show that the greatest decay rates occur in the finer fractions. Runs 3 and 4 show that the short-lived fraction appears only with the power on. Both of these findings are consistent with the theory that part of the activity is due to fission gases decaying in flight, the active daughter of which deposit on surfaces of particles. Some of these daughter products may also deposit directly on the slides.

Radio-autographs were made of runs #1 and #2, stages a-d. Eastman's Industrial Type K film was used at 72 hour exposures. Detectable discrete centers of darkening were counted as particles. The results appear in Table VIII.

Table VIII  
Radio-Autographs of Cascade Impactor Slides

Sample	Mean Diameter of particles (microns)	Beta and Gamma		Number of Particles Detected	c/m/Particle
		c/m (10/5)	c/m (10/8)		
1A	6	1372	1096	140	7.8
1B	2	272	124	56	2.2
1C	1	293	81	41	2.0
1D	0.7	176	46	15	3.1
2A*	6	987	531	44	12.1
2B	2	625	154	73	11.6
2C	1	485	88	88	1.0
2D	0.7	309	48	38	1.3

\* This slide was broken, and impingement pattern was irregular. It seems that the air stream was distorted, making A and B fraction inaccurate.

These radio-autographs showed a diffuse background in addition to discrete spots. Consequently, c/m/particle as shown in the table is larger than the true value. Radio-autographs of Run #1, A-D are shown in Figure XV.

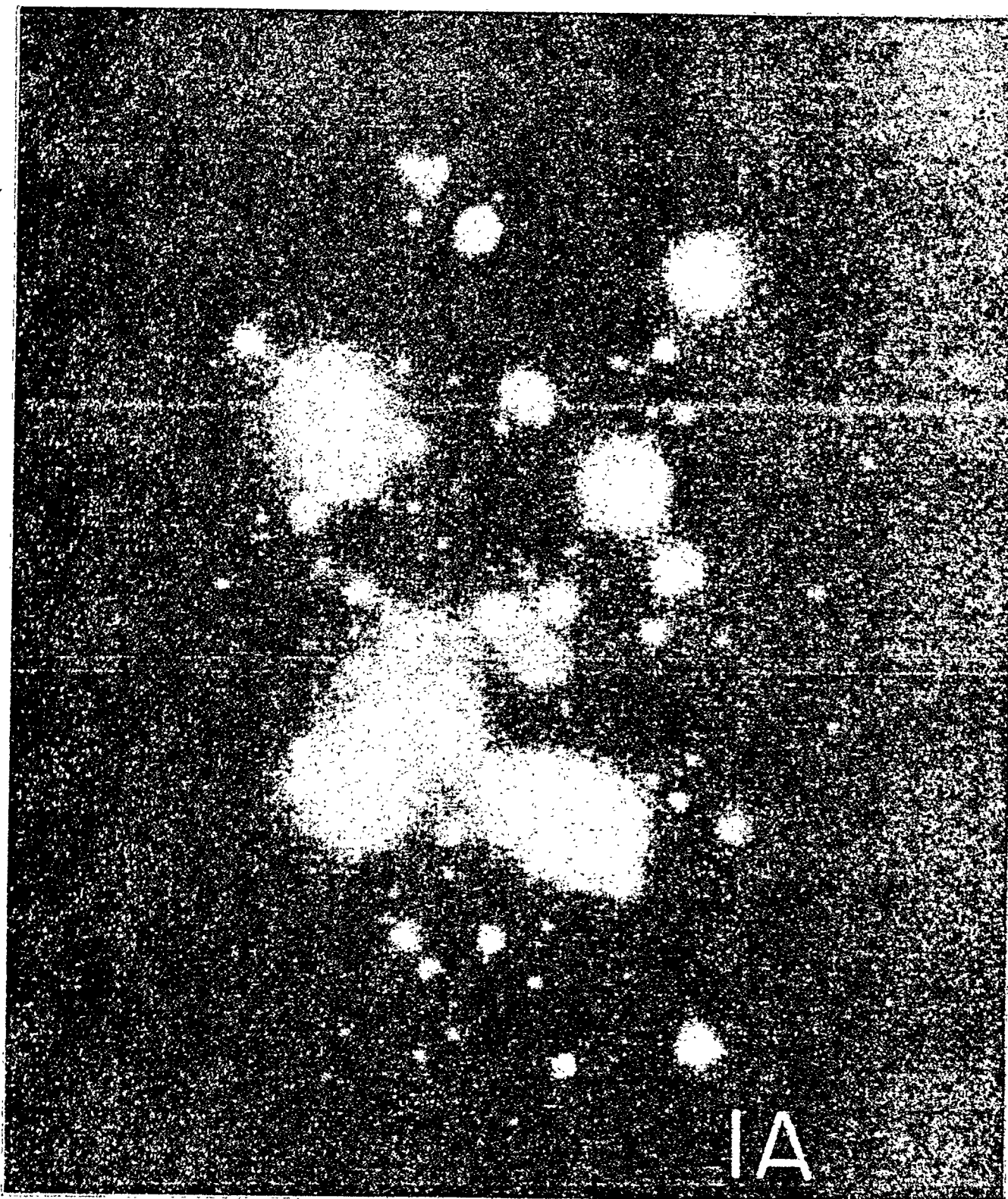


FIGURE XVa

RUN #1  
CASCADE IMPACTOR SAMPLE A



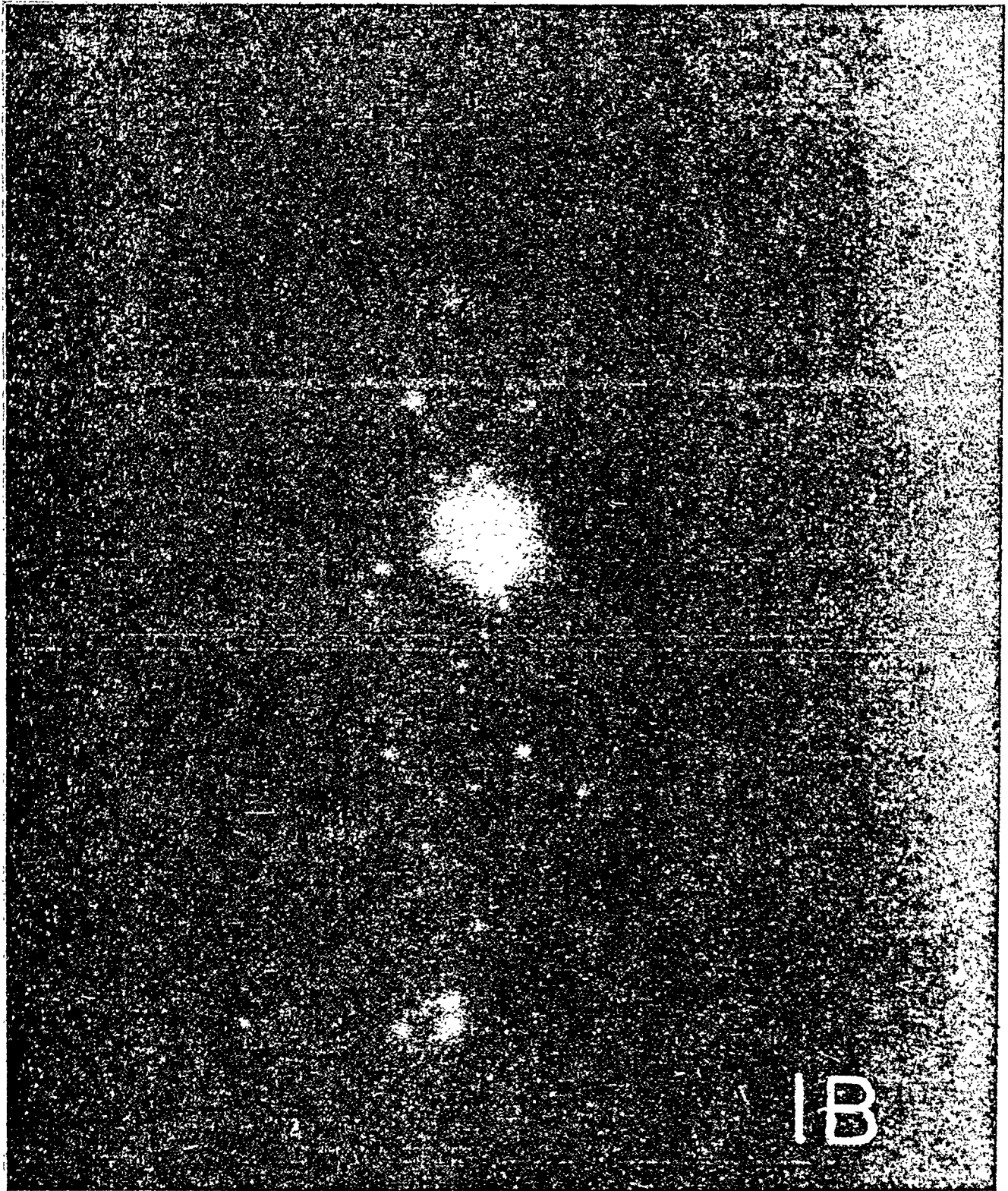


FIGURE XVb  
RUN #1  
CASCADE IMPACTOR SAMPLE B

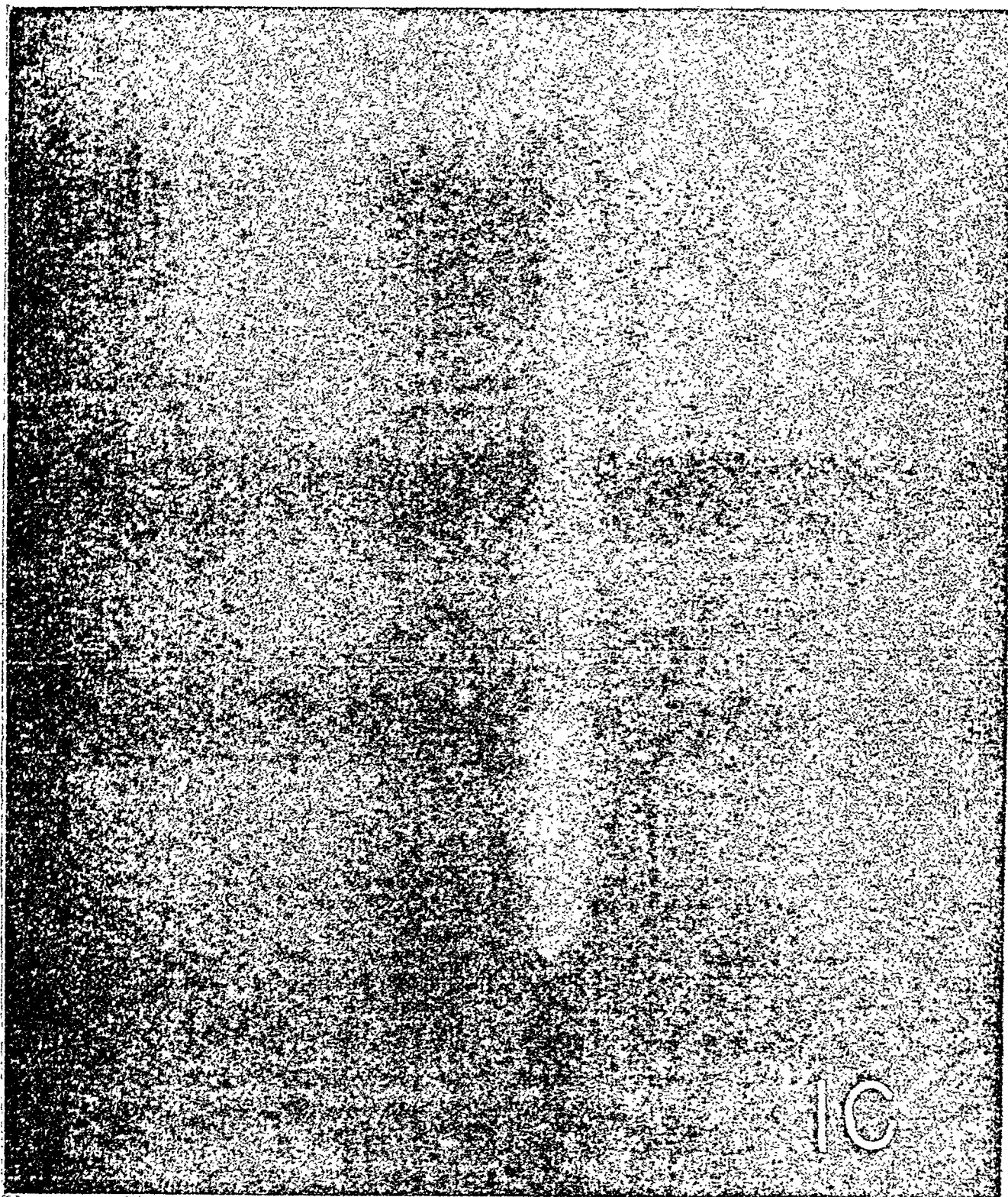


FIGURE XVc

RUN #1  
CASCADE IMPACTOR SAMPLE C

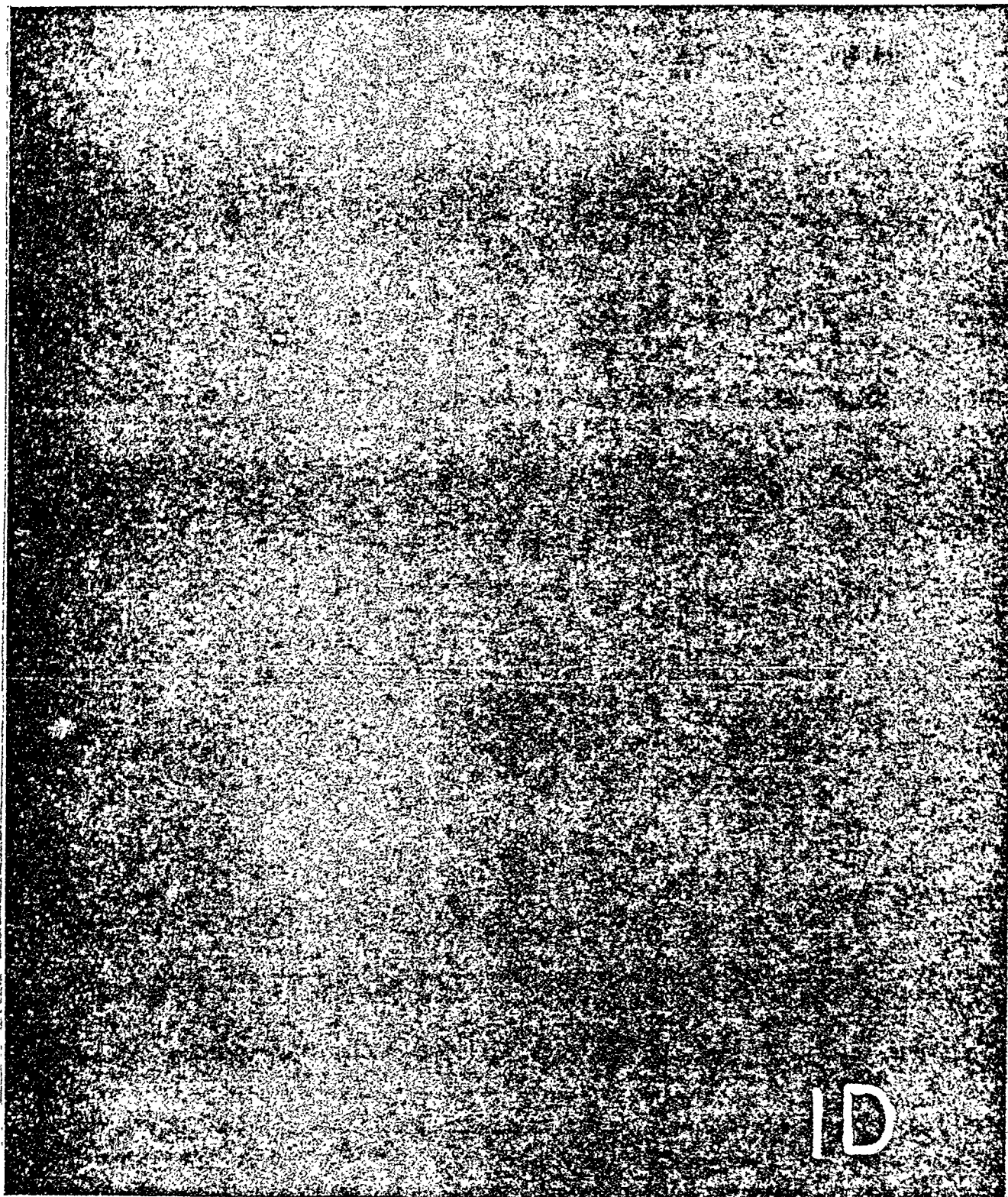


FIGURE Xvd

RUN #1  
CASCADE IMPACTOR SAMPLE D

A report from Rochester gave some figures on particle size distribution based on these cascade impactor samples. The samples were analyzed for uranium content by the fluorophotometric method. It was assumed that all uranium detected was in the form of  $UO_2$ . It was further assumed, for the basis of the calculations, that particle densities in each size range do not differ from the true density and that all particles approximate spheres. Results of the analyses and calculations appear in Table IX.

The figures in column six were derived from the weighted average of samples #1 and 2, calculated on the basis of 110,000 cfm (full air flow - 2 fans), or  $4.49 \times 10^5 m^3/day$ .

These figures are perhaps not as much of a cause for immediate concern as appears at first glance. Dust studies have indicated that particles less than  $0.3 \mu$  become permanent atmospheric impurities, and that particles less than  $0.1 \mu$  act as gas molecules. Even a  $4 \mu$  particle of  $UO_2$  takes 184 minutes to fall 200' (Stokes law,  $T = 70^\circ F$ ), and the average wind velocity in this area is  $\sim 6$  mph (6.2 for 1948), so that the average distance traveled by a particle of this size and nature is  $\sim 18$  miles. Obviously this is not accurate, since the terrain is not level, but, on the other hand, the orifice velocity and temperature

Table IX

Particle Size and Count Analysis of Stack Samples in Terms of  $UO_2$

Size Range (u)	Particles/10 m <sup>3</sup>				Particle/Day (Normal Operations) Based on Av. (#1 and 2)
	Sample #1	Sample #2	Sample #3	Sample #4	
0 - 0.5	$1.4 \times 10^7$	$1.1 \times 10^7$	$1.0 \times 10^7$	$5.3 \times 10^7$	$5.38 \times 10^{12}$
0.5 - 1.0	$1.1 \times 10^6$	$6.6 \times 10^5$	$7.5 \times 10^5$	$1.8 \times 10^6$	$3.64 \times 10^{11}$
1.0 - 2.0	$1.7 \times 10^5$	$9.7 \times 10^4$	$1.2 \times 10^5$	$2.2 \times 10^5$	$5.44 \times 10^{10}$
2.0 - 4.0	$1.5 \times 10^4$	$8.5 \times 10^3$	$9.9 \times 10^3$	$2.1 \times 10^4$	$4.80 \times 10^9$
4.0 - 6.0	900	436	555	1600	$2.65 \times 10^8$
6.0 - 8.0	140	42	66	270	$3.37 \times 10^7$
8.0 - 10.0	26	15	12	64	$8.5 \times 10^6$
10.0	2.4	0.83	1.3	12	$6.3 \times 10^5$

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differential of the effluent air both tend to carry the entrained particles to a greater height. This effect is enhanced during conditions of low wind velocity when the particles would otherwise fall closer. At such distances the dilution factor is quite high.

It is not intended to imply that this is not an undesirable condition since long term operation would allow cumulative effects. Besides, the larger particles, which do fall on the immediate plant area, while somewhat large to be considered breathable, are subject to fragmentation and elevation by traffic and wind, and these effects occur near the ground.

The evaluation of size and frequency of occurrence of such particles in the breathing zone of personnel in the area is a more difficult problem. Filter media do not differentiate size fractions, and the cascade impactor, with its capacity of 0.5 cfm, does not lend itself to sampling an atmosphere where the expectancy of active particle incidence in this size range is on the order of one to two per 1000 ft<sup>3</sup>.

Of more immediate interest was the finding that 105 building was highly contaminated with fine radioactive particulates which were easily airborne. C.J. Borkowski found up to 75 particles/1000 ft<sup>3</sup> of air in sampling with an Airway, and radioautographing without the bag

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as absorber. Our Airway monitoring program showed that a machine set on top of the reactor gave the highest particle count of any location, and that this was 4 to 10 times the average collection rate of the other ten. Borkowski also tested several surfaces in the 105 building with film wrapped in a double envelope of 1 mil Al foil. He found that surfaces were highly contaminated with particles with activities ranging from 10 d/m to several thousand.

R.L. Clark, of Health Physics, investigated the problem of locating the source of this contamination. He found that, although the reactor is maintained at a negative air pressure, any operation involving the movement of objects into and out of the reactor caused a mechanical transfer of particles out of the reactor. The operations monitored included:

1. Scanner plug removal
  2. Control rod operation
  3. Shim rod operation
  4. Removal of experimental samples from front face
  5. Slug discharge and recharge
  6. Removal of sulfur cans
  7. Pulling of isotope stringers
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Results of these tests varied widely and some of the operations released thousands of particles which were picked up by an Airway covering them. Apparatus was developed to sweep stringers with a combination of a mechanical brush and an air stream which then passes through a filter. Push-rods used in slug and stringer manipulation were wiped with a damp cloth as they were drawn out. There was also a general clean-up of 105 building.

Air sampling tests on January 7-13, 1949 showed a 90% reduction in particle count. Other tests indicate a 90 to 95% improvement.

The film techniques used by Borkowski for determining surface contamination on flat surfaces in and around 105 building was tried for the general plant area. The Al foil covered films were laid out for 48 hours, October 16-18, covered with 1/2 inch plywood boards to shield them from weather and sun. A heavy rain (1.65 inches) fell during this period and many of the films sustained water damage. Those not damaged showed results of poor geometry in many cases, and particles of small activity were not detected. Figures XVI and XVII illustrate the kind of results obtained. The film shown in Figure XVI was obtained on the roof of the fan-house (building 115). The effects of small particles that have settled between the gravel of the roof can be seen. Figure XVII depicts the radioautograph obtained on a grassy area ~ 200' E of the 105 stack.

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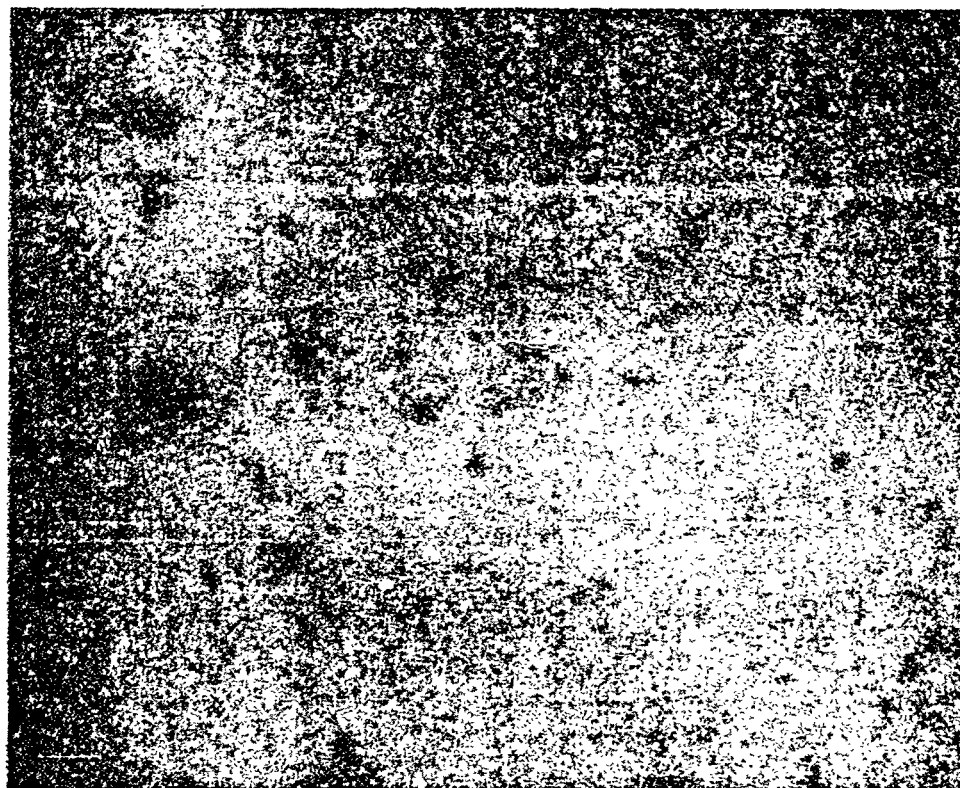


FIGURE XVI  
ROOF 115 FAN HOUSE

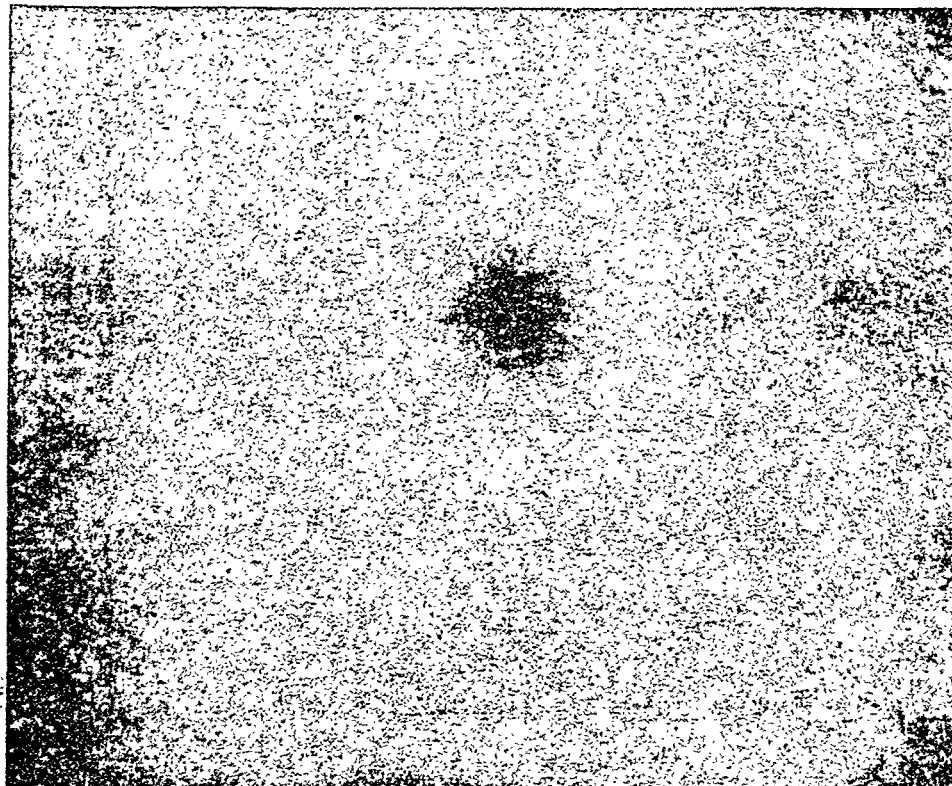


FIGURE XVII  
GRASSY AREA 200' EAST OF 105 STACK

With a few refinements the method has merit for determining ground contamination. If the area to be covered is flattened to provide good geometry, and a shallow drainage trench provided around the small area covered to prevent water damage, the results should be quite representative of true conditions. The experiment, with these improvements, was not repeated because the grassing of the plant site, with the covering of the original surface with inches of top-soil, proceeded rapidly at this time, so that coverage in the area of greatest interest could not be obtained.

During the period, 9-17-48 to 11-17-48, a total of 767 nose swab samples were taken at the dispensary from a selected number of people upon completion of the day's work. The cotton swabs were sliced open, counted in a standard beta-gamma counter at 10% geometry, and were then radio-autographed for 24 hours. By this technique particles were detected on 66 samples out of the entire group, or 8.6%. The number of individuals reporting daily for swabbing fluctuated widely from a high of 64 on one day to a low of 8. The daily average for the last five weeks was approximately 14.

Particles were detected on a greater percentage of samples during the period 9-21-48 to 10-8-48 than any other time, but during this period the greatest number of people reported to the dispensary

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for swabbings. Relatively few particles have been detected since 10-8-48, but during this period the number of people reporting daily has fallen off markedly.

Of the total of 767 swabs, 138 were contributed by a group working in Building 105, with a particle incidence of 12.3%, as compared with an incidence of 7.8% in the remaining 629 swab samples. As previously noted, air collections taken within 105 have indicated a greater density of particulates than any other location.

These results gave further evidence of the presence of air-borne radioactive particulates. The figures do not supply a good measure of the extent of this type contamination, as it is felt that negative results are not conclusive.

Because of very high particulate air count directly traceable to the November RaLa run, it was decided that this operation receive closer study. Consideration of the information available produced some interesting features. Technical Division sampling the D-stack effluent estimated that ~ 15 curies of activity were discharged with the cell ventilating air. The activity reached a maximum during the final evaporations and removal of the product, which took place early on the morning of Sunday, November 21. An inversion with low wind

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velocity existed at the time, and caused the activity to spread and be retained in the valley. Outside constant air monitors, assuming that particulate activity followed the same pattern as total activity, showed that  $\sim 70\%$  of all contamination was collected between 3:00 AM and 7:00 AM. During this period density was  $\sim 1$  radioactive particle ( $\geq 7$  d/m) per  $\text{ft}^3$  of air.

On the basis of these findings, a temporary filter similar in plan to the reactor filter was designed and installed in the cell vent duct. Consequently, the January RaLa run presented a different picture. A discharge of  $\sim 0.1$  curie of activity was estimated by the Technical Division.

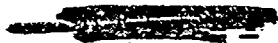
For special study of this run the D-stack was monitored by means of a mobile constant air monitor sampling the stack effluent from 5' below the top inside by means of a 1" rubber hose. Filters were changed every two hours during the first stages of the cycle. The peak of particulate activity did not coincide with the peak of total activity. Peak of particle density occurred during transfer operations from B-6 to B-19. Total activity also showed a high value at this time, but in addition showed a peak at the end of the evaporation cycle which had no counterpart in particle count. The point of greatest significance is

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the fact that particle density was greater on the E platform of 706-D, as detected with both a filtron and a USPHS filter, than in the stack itself. This indicated that particulates were coming through the building rather than the stack.

Two of the large USPHS filters were transferred off the plant area for this run. One of them, placed 2.5 miles west of the plant, showed only 0.003 particles/1000 ft<sup>3</sup>. The other, at Scarboro School, 6 miles ENE of the plant, showed 0.408 particles/1000 ft<sup>3</sup>. Since the wind was westerly throughout most of the run this pattern is as expected. It is significant that the latter value is higher than the averages of particle densities detected at the plant area when the immediate vicinity of 706-D building is excepted.

Both this phenomenon and the fact that a considerable number of particles seem to pass through the filter indicate that the contaminant is in the form of extremely small particles, or in droplets or mist which saturate the filter and are blown off again from the exit side. The extreme minuteness of the particles was also indicated in a subsequent observation that particulates seem to spread by diffusion during removal of the product for shipment, even though there is an air draft into the cell when this operation is performed.



Discussion:

While the subject of this investigation was a variation of a dust problem, little in the previously developed dust study techniques was directly applicable. The chief point of difference was that the order of magnitude of contamination or count density levels was totally different. Dusts containing silica are considered to reach a dangerous concentration at levels of 5 to 50 million particles per ft<sup>3</sup> depending on free silica content, whereas, concentrations of radioactive dust, except occasionally, were on the order of one particle per 1000 ft<sup>3</sup> - a factor of 10<sup>10</sup>. Normally "clean" air may easily contain 10<sup>5</sup> dust particles per ft<sup>3</sup> in suspension. As a result, the isolation of a radioactive particle of micron range is next to impossible even after capture by filter or other medium.

Many of the methods used were comparatively gross, but we were faced with the problem of collecting a mass of information in a short time. There was usually not time to develop or refine techniques since many of the effects studied were transitory. Construction of filters, grassing and paving of the plant area, and general clean-up operations were in progress during the course of the investigation, so that a continuation of an already established procedure gave a better comparison of conditions than would have any new technique which might be an improvement as regards an approach to an absolute evaluation of level of contamination.

The assumption that the sedimentation frame collections, though actually consisting mainly of particles too large to cause a breathing hazard, were a measure of air contamination originating in the reactor was justified by the findings of the cascade impactor sampling of the reactor duct. Even this study did not, however, give a measure of the concentration of particles of breathable size from this source, in the working zone of the plant. We can, nevertheless, reasonably assume that the degree of improvement in the lower size range closely follows the diminution shown by frame collections. Judged by this standard, the filter-house is found to be doing a satisfactory job of reactor coolant air decontamination.

Results on the major isotope separation processes are neither as favorable nor as clear-cut. There was a great improvement shown in subsequent runs over the November RaLa run, but it has not been established that the November run was characteristic. It is noticeable that dissolver operations of all types, the off-gases from all of which are vented through the 200 ft 205 stack, were very seldom noted as contributors to local air contamination, while the gaseous or airborne wastes from the latter part of the separation cycles of the isotope processing areas, vented through short stacks, were almost always significant contributors. It may be hoped that with the use of the new 250 ft isotope stack this condition will be remedied, especially since the wastes will be passed through purifiers.




The nature of the contaminant from the isotope areas is not known as well as that of the reactor-produced contaminants. It is known that differences exist. A large fraction of the chemically-produced activity is water-soluble. A close evaluation of this factor is rendered difficult by the co-existence of other contamination in a vapor or fume state which appears in a radio-autograph as a diffuse darkening. Large particles either do not exist or are very few, as shown by the high ratio of filter-captured particles to what appears in the sedimentation frames. It has not been determined whether the centers of contamination which have been detected as "particles" existed in air in the form of solids or liquid droplets. There is indirect or presumptive evidence that the "particles" are extremely minute, and of high specific activity. The average half-life is short compared to that of the reactor produced particles. Furthermore, the contaminants originating in the chemical operations are free of  $\alpha$ -emitters.

Further study on the size distribution and the physical and chemical nature of the chemically produced particles is planned. The work will be done by the University of Rochester. Such information is needed in any attempt at evaluating the potential health hazard due to these particles.

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At the present time information is insufficient to judge what hazard, if any, particulates, such as described, introduce into the environment of plant personnel and inhabitants of the vicinity of the plant. Previous dust studies have established that dust most likely to reach, and be retained in, the alveoli of the lungs comprises the size range of  $\sim 0.5$  to  $5.0 \mu$ . The history of particles  $> 10 \mu$  which may get into the bronchial tract, but not reach the alveoli, has not been studied, as they are not significant from the point of view of silicosis hazard. It is conceivable that an insoluble particle of high specific activity may become lodged in the bronchial tract and cause a high degree of local irradiation. The effects of irradiation on the protective mechanism is unknown. There is also the possibility that small insoluble radioactive particles, each by itself being innocuous, may be concentrated by phagocytic action to the point where the accumulated radiation level may be damaging.

The final product of the RaLa operation is  $\text{Ba}(\text{NO}_3)_2$ . If this product is pure and dry, its specific activity is  $0.13 \mu\text{c}/\mu^3$ , so that, while free of alpha-emitters, and also soluble, particles small enough to remain in relatively permanent suspension ( $\sim 0.3 \mu$ ) may contain  $10 \text{ m} \mu\text{c}/\text{particle}$ .



A final evaluation of the actual hazard due to radioactive particulate air contamination will entail a long-range program of pathological study. The level of gross air contamination is well below the accepted tolerance of  $8.5 \times 10^{-8} \mu\text{c/cc}$  (based on  $\text{I}^{131}$ ), except for occasional short periods in the isotope separation area. Although the consensus of opinion is that small isolated areas of tissue are more radio-resistant than are large areas, the safety factor introduced thereby is, at best, a matter of conjecture. After the preliminary study, previously mentioned, has established the size distribution and nature of the various types of particles produced in the different operations, pathological studies will be required to trace the history and influence of each type of particle in lung tissue.

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